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Environmental Testing of Fine Interconnections Ink Jet-Printed on Flexible Organic Substrates

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Abstract. *This article introduces ink jet-printed nanoparticle-based silver ink for integrated circuit interconnections. The environmental reliability of these interconnections at varying temperatures has not been exhaustively reported nor their electrical resistance tested at constant humidity. The authors ran two environmental tests to examine the reliability of ink jet-printed interconnections: 1000 h at 85°C at 85% relative humidity (RH) to evaluate the effects of humidity and 1000 cycles at −40°C–125°C for the effects of temperature based on common industrial test standards. Results showed no silver degradation, and the electrical resistance fluctuated ~3–6% for humidity and ~5–6% for temperature. The results were mathematically modeled to understand the acceleration factor in the humidity test in the chosen user condition, i.e., at 50% RH at room temperature and at time-to-fail in the temperature cycling. The ink jet-printed interconnections showed good reliability. © 2009 Society for Imaging Science and Technology.*
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INTRODUCTION

Compared to traditional photolithographic processing, the ink jet printing of conductive interconnections offers several advantages such as maskless production, savings in material and energy, fewer process steps, hardly any waste liquid during manufacture, low-cost, and reduced environmental impacts.[1] The fast processing of interconnections represents one of the benefits of transferring the technology to mass manufacture.

Nanotechnology has made nanoparticle-based inorganic inks available for printed electronics to expedite manufacture and to save energy in producing interconnections of electronic circuitry without compromising their high electrical performance requirements. The growing demand of such circuitry in the electronic packaging industry has dramatically increased their package density and functionality. The main challenge is to define the printing steps to produce fine interconnection lines of high electrical performance without compromising their reliability.

We tested ink jet-printed silver interconnections in several climatic environments according to well-known and widely applied industrial electronics standards. In addition, we calculated the interconnections' acceleration factor in a humidity test to define their technological life and predicted their time-to-fail value in chosen temperature cycling test conditions.

NPS INK AND FLEXIBLE ORGANIC SUBSTRATES

In recent years, exploiting the benefits in printed electronics of formulating printing ink from inorganic nanoparticles has been of increasing interest. Nanoparticle-based inks offer several advantages in printed electronics, such as low-temperature sintering, low-viscosity requirements, and suitable particle size for ink jet printheads. The sintering temperature can be lowered because of the "quantum size effect,"[2] also enabling printing on flexible temperature-sensitive substrates. In addition, a wide range of metal-containing (e.g., silver, gold, nickel, and copper) nanoparticles are available for bulk materials, making it easy to formulate a rheologically suitable ink for a particular ink jet printing medium. The ink's physicochemical properties, such as viscosity and surface tension, are its most important parameters directly affecting the printing process and the quality of the printed component. The theoretical requirements of ink jet ink for viscosity and surface tension are in the range of 2–30 mPas and 25–35 dynes/cm, respectively. Nanoparticle-based ink must also show Newtonian behavior to have reliable sheer stress under variable temperature and pressure.

During printing, the printhead temperature may vary because of the temperature changes on the printing *XY* table. In addition, the short distance, usually <1 mm, between printhead and substrate may alter the printhead temperature during long printing runs and cause drying at the nozzle. On the other hand, the selected ink material should be less than 100 nm in particle size, and the nanoparticles should be uniform and homogeneously distributed in the solvent. Their dispersion in ink is important to avoid aggregation, which may increase the ink's viscosity during printing and lead to unwanted clogging of printhead nozzles.[3]

As mentioned above, nanoparticle stability in ink is the challenging factor in printing and storage and must be controlled with a liquid vehicle. In this study, ethylene glycol, glycerol, and an ethanol liquid mixture were used to control

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Table I. Properties of NPS ink.

	Before sintering process	After sintering process
Particle size (nm)	30–50	
Metal content (wt %)	20	
Viscosity (mPa s)	12–14	
Specific resistance ($\mu\Omega$ cm)		5
Thickness (μm)		$\sim$2–3

Table II. Properties of tested flexible organic substrates.

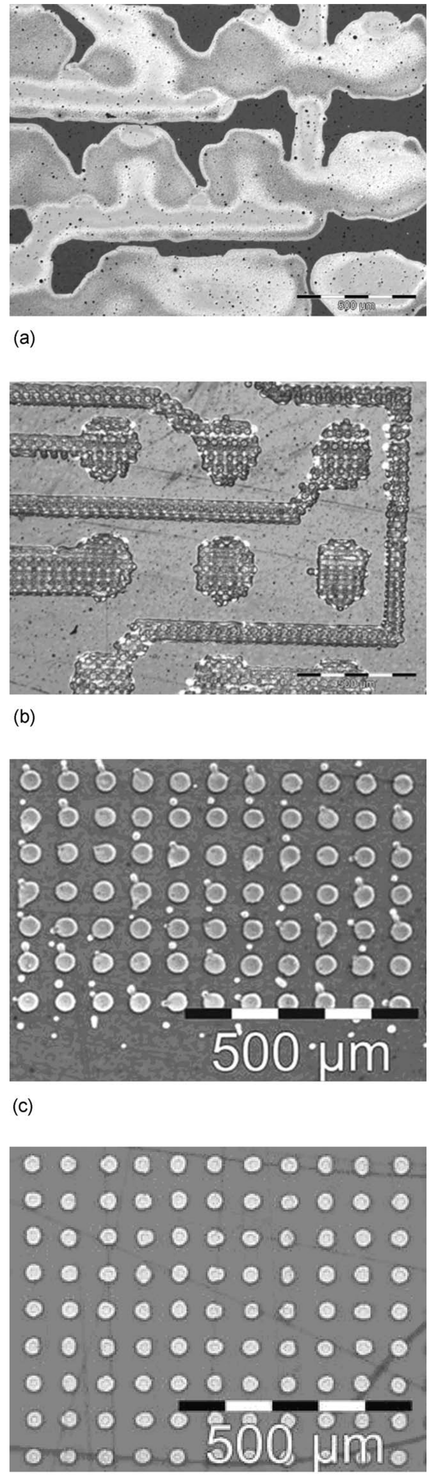
	PEN	LCP	PI
Thermal expansion (CTE) (ppm/°C)	18–20	17	20
Tensile strength (MPa)	250	294	139–231
Melting point (°C)	270	310	
Dielectric constant	2.90	2.85	3.50
Moisture absorption (%)	0.40	0.04	1.80
Substrate thickness (μm)	50	50	50

nanoparticle stability. Table I describes the properties of the nanoparticle-based silver (NPS) ink.

Because NPS ink must be printed on a suitable organic substrate, substrates must be selected carefully in terms of their sintering temperature and durability. In addition, organic substrates' engineering properties, such as dielectric constant, moisture absorption, and coefficient of thermal expansion (CTE), are important for several electronics applications, including e-paper, organic radio frequency identification (RFID) tags, and flexible organic light emitting displays.[1] In this study, we tested polyethylene naphthalate (PEN), a liquid crystal polymer (LCP), and Kapton polyimide (PI); the properties of these substrates are given in Table II.

PRINTING PROCESS AND ITS CHALLENGES

Direct writing from a computer-assisted design image file with a piezotype ink jet printer has substantial advantages over current electronic manufacturing processes, i.e., photolithography. Process steps can be dramatically cut in surface cleaning and modification, digital printing, and the final curing of NPS ink, and the whole process can be repeated with a dielectric layer and other NPS ink combinations. In our study, before printing, substrate surfaces were cleaned of dust and possible contamination with isopropanol and then treated with ultraviolet irradiation and ozone for 5 min. The surface modification was finalized with an electronic coating liquid consisting of low-viscosity fluorosilane polymer in a hydrofluoroether solvent. This process step enables the control of the droplet size on a flexible organic substrate and decrease in the linewidth to produce fine interconnection lines (see Figure 1).



(d)

Figure 1. (a) Bulking of NPS ink droplets, (b) droplet control by masking the printing files, (c) satellite droplets forming during printing, and (d) their effect controlled by adequate pulse voltage and ink with a high metal content.

Several challenges may arise during printing. One major problem with industrial ink jet printers is the bulking of the droplets. However, the problem can be solved by masking the printing files, selecting the right waveform, by controlling the temperature of the printhead, or by some combination of the above. Another problem is satellite droplets, which occur mostly because of the high pulse voltage and low-metal content of NPS ink. Pulse voltage can be lowered according to the ink's specific wave form. Ink viscosity should always remain in the range of industrial printheads, and the printer operator should be aware that very low-viscosity ink, e.g., <1 mPa s, might compound the effect of satellite droplets. In this study, we set the printhead temperature at 40°C and 60°C for the *XY* printing table during printing. Droplet bulking was controlled by surface treatment, surface modification, and masking the printing patterns. An appropriate wave form was used for the NPS ink together with 14 V printhead piezovoltage to minimize satellite droplets. Fig. 1(a) shows bulking of NPS ink droplets; Fig. 1(b) illustrates the droplet control by masking the printing files. Fig. 1(c) shows satellite droplets which form during printing, and Fig 1(d) illustrates their effect controlled by an appropriate pulse voltage and the use of an ink with a high metal content.

It was important to select the right printing resolution and to increase the number of NPS ink layers to make interconnections more reliable at varying temperatures and humidity.[1] In addition, for multilayer printing with low-metal-content NPS ink, the surface of each ink jet-printed layer had to be of good quality. Four-layer printing with 600 dpi was compared to single-layer printing with 1550 dpi according to the reliability tests described in our preliminary report.[1] The former was considered suitable for printing fine interconnections when the repetition of a printing layer was intended to directly effect sheet resistance at the applied printing resolution for producing an RFID antenna or interconnections.[4] Consequently, we managed to print interconnections of linewidth 190 μm and thickness $\sim 2-3$ μm.

ENVIRONMENTAL TESTS OF INK JET-PRINTED INTERCONNECTIONS

Environmental reliability testing of ink jet-printed interconnections is important to verify and confirm the quality of printed structures before the product is marketed. In addition, such testing can help define the necessary requirements for quality assurance.[5] In everyday use, electronic devices are subjected to various environmental conditions. They are exposed to sunlight, ventilation, low or high temperature variants with humidity, and vibration, which may cause partial or total failure of a subsystem. The failure of a mobile device may thus serve as an example. In printable electronics, the reliability issue has not been exhaustively explored, and industrial standards are still required even for several extensively researched applications such as RFID tags, passive components, transistors, interconnections, and display back panel wiring. In fact, the Institute of Electrical and Electronics Engineers, Inc. has recently suggested several standards for electrical requirements of organic transistors; standards that could be viewed as the organization's interest in this particular issue.[6,7]

We ran several tests to determine the major environmental effects on ink jet-printed interconnections on flexible organic substrates. As we know, moisture invades porous substances and causes oxidation from conductance and corrosion between conductive materials in humidity tests. Moreover, a thermal shock may permanently alter electrical performance, and a sudden overloading of materials may cause cracking and mechanical failure.[5] In this study, the following test conditions were applied to determine problems related to humidity and temperature variation in ink jet-printed silver structures:[8,9]

(1) 85°C and 85% relative humidity (RH) applied for 1000 h to test effects of humidity. According to the supplier, the fluctuation in the humidity chamber was ±0.3°C and ± 2.5% RH. The samples were measured for electrical performance at every 250 h.
(2) −40°C–125°C temperature cycling was used for 1000 cycles of 30 min each. The samples were measured for electrical performance at every 250 cycles.

The above test conditions represent extreme user conditions for a mobile device or internal electronic components on a printed wiring board (PWB) and bear directly on understanding the life cycle of unprotected interconnections. Both test conditions were selected without compromising the other electronic components—electronic assemblies and packages/components—working with the interconnections possibly sustaining nonfuctionality, mechanical damage, corrosion/electromigration, or surface degradation of PWB.[10]

Resistance fluctuation was measured with a Greek cross test structure adapted from the semiconductor industry. The Greek cross circuit has four pads connected to each other at equal lengths and linewidth. The symmetrical structure compensates for contact resistance and provides reliable results, extensively, discussed in the literature.[1]

However, the sintering condition of the selected NPS ink may affect the reliability results of ink jet-printed samples. In this study, we used 220°C for 60 min to sinter the ink jet-printed fine interconnection lines. The sintering temperature of the silver ink played an important role when we reviewed the criteria for using PI, PEN, and LCP substrates.

RESULTS AND DISCUSSION

The reliability of ink jet-printed NPS on flexible organic substrates was evaluated in the temperature cycling and humidity tests. Change in the resistance value was measured after each 250 cycles of temperature cycling and 250 h of humidity testing. Our results show that the electrical performance fluctuated at varying temperatures and at constant humidity in a similar fashion. In temperature cycling, resistance values were measured and fluctuation was observed as between ~ 5 and 6%, depending on the tested substrate [Figure 2(a)]. The results also show a trend similar to that previously reported,[11] where the stability of silver particle-based conductors was measured in a thermal cycling test up to 1000 cycles at −40°C–+85°C. A change was detected in the resistor values of tracks varying in width from

100–500 μm with conductors of width 200 μm printed in a continuous reel-to-reel rotary screen process for printed antennas in smart label/card applications, turning out to be the most stable in their resistor values.

Our CTE values for the ink jet-printed thin film layer[12,13] and the flexible organic substrates were close enough to each other for mechanical failures, such as voids, major-cracks, or line fractures, not to be detected after the temperature cycling test. However, we detected microcracks on the ink jet-printed NPS film surface on the PI substrate and described them in our previous paper.[1] Microcracks may trigger wider cracks and result in electrical nonfuctionality of the interconnection lines. Several material parameters, such as CTE, elongation and the physical dimensional change ratio of the substrate, and the sintering profile of the ink, play an important, direct, or indirect role in the temperature cycling test.

In contrast, resistance values fluctuated in the humidity test between ∼3 and 6%, depending on the tested substrate type [Fig. 2(b)]. The spreading of the droplet on the organic substrate directly affects the quality of the ink jet-printed interconnection line in terms of electrical conductivity, linewidth, and moisture penetration to nanoscale porosity at constant humidity in the operational environment. In addition, substrate properties, such as the moisture absorption rate and the physical dimensional change ratio at the selected 85% RH affected the humidity results.

In this study, we standardized as many printing and sintering constants as possible, such as printing resolution,

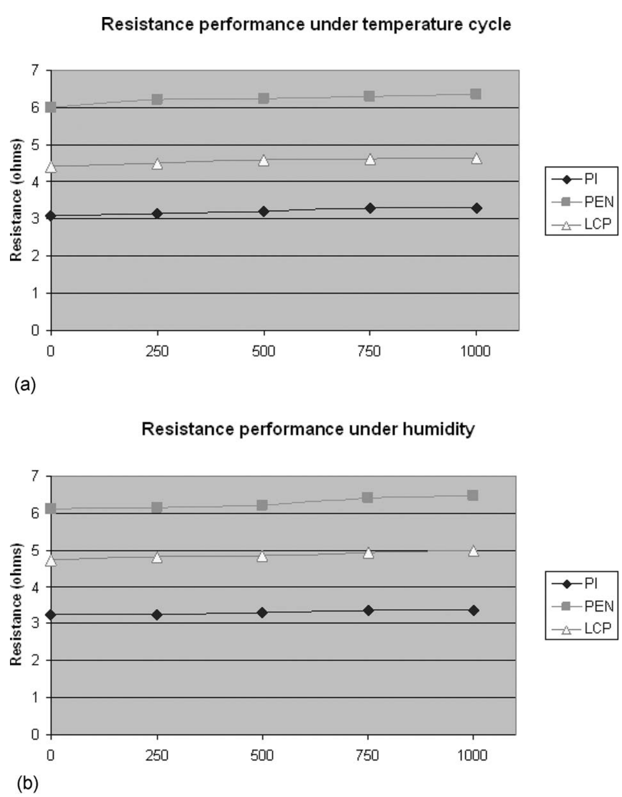


Figure 2. Changes in resistance performance during (a) temperature cycling and (b) humidity tests.

substrate surface cleaning and modification, sintering temperature, and durability of the NPS ink. Thus we could relate test results directly to the substrate-ink material interaction and its properties. To analyze the role of the material and test condition for a certain application, two mathematical parameters in the test results must be clarified: A_f (acceleration factor) for humidity and the prediction of t_f (time-to-fail) value. Several physical models, e.g., Arrhenius, Eyring, Lawson, Memis, and the Coffin Manson, have been suggested for the analysis of environmental reliability such as temperature cycling and humidity.[10] They are suitable for mathematical modeling of ink jet-printed interconnections based on accumulated reliability data and reflect well enough the role of materials and structures in test conditions. Mathematical A_f analysis, however, can be performed only if a certain type of electronic application was selected. We speculated that for humidity analysis, the operating condition of most electronic devices is likely to be 50% RH and constant room temperature (25°C).

We used the Coffin Manson equation to calculate A_f.[14] The same model has also been used to calculate the fatigue life of joints between a silicon chip and ink jet-printed NPS thermal interface material[15] and considered as a relevant model to apply to our study. The model suggests that A_f is defined as

$$A_f = \left(\frac{RH_1}{RH_2}\right)^n \exp\left[\frac{E_a}{k}\left(\frac{1}{T_1} - \frac{1}{T_2}\right)\right], \tag{1}$$

where RH_1 = the relative humidity of the application, RH_2 = the relative humidity in the humidity test, E_a = the electronic activation energy in eV (electron voltage), k = Boltzmann's constant (8.617×10^{-5} eV/°K), T_1 = the temperature (°K) of the application, and T_2 = the temperature (°K) in the humidity test.

However, because we could not measure the activation energy of our NPS ink jet-printed interconnections on the organic substrate, we used activation energy (typically from 0.77 to 0.90) and exponent n (typically from −2.5 to −3) values from plastic-encapsulated microcircuits,[14,16–18] where interconnections to the die can be printed with NPS.[19] We therefore selected $E_a = 0.8$ eV and $n = -2.75$ for quality parameters and measured A_f as ∼14.98. The acceleration factor defines the technological life of the ink jet-printed structure for the selected condition as

$$1000 \text{ h(test duration)} \times 14.98$$
$$= 14{,}980 \text{ h(technological life time)} = 1.71 \text{ yr}.$$

However, we must also consider that the life cycle calculated for unprotected ink jet-printed structures which also might be even more reliable and have longer technological life when the interconnections are protection coated. For example, the traditional conformal coating was used to protect PWB and insulate wire or capsulated with plastic encapsulant which might also increase the overall reliability of the interconnections.

On the other hand, the following temperature cycling test model was used to estimate the t_f value:[10]

$$t_f = A \cdot e^{b/\Delta T}, \tag{2}$$

where ΔT=the change in temperature (set minimum and maximum temperatures in the test), and A and b are constants.

Values for A and b represent NPS ink and substrate material properties, i.e., metal content, substrate dimensional change ratio, and CTE, which also consider the constant printing and sintering parameters discussed above. Constant A correlates with the CTE value difference between the ink jet-printed NPS layer and the substrate and also takes into account dimensional change. At 50% RH, the dimensional change ratios for LCP, PEN, and PI were 0.02%, 0.05%, and 0.12%, respectively.[20] Constant b is the temperature change exponent with a typical value of about 2.[21] Calculation yielded the t_f values of 5940, 1833, and 1205 cycles for LCP, PEN, and PI, respectively. However, these are simulated results and require further field testing.

CONCLUSIONS

NPS ink was ink jet-printed on flexible organic substrates and tested for its interconnecting performance in temperature cycling and humidity tests. The fluctuation of electrical resistance versus cycles in the temperature cycling test and versus hours in the humidity test were evaluated. The results showed no significant fluctuation in the resistance of the samples. The resistance fluctuated $\sim$3–6% for humidity and $\sim$5–6% for temperature cycling. The results were also modeled with several related mathematical models to predict the technological life of the samples at 50% RH at room temperature and their time-to-fail in the chosen temperature cycling condition. The theoretical results may vary according to the printing and sintering profile variables and the humidity and temperature inconstant in the operating conditions. However, our NPS ink jet-printed interconnections showed a good degree of reliability in the environmental tests.

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