Printable electronics: towards materials development and device fabrication

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Abstract

Purpose – There has been increasing interest in the development of printable electronics to meet the growing demand for low-cost, large-area, miniaturized, flexible and lightweight devices. The purpose of this paper is to discuss the electronic applications of novel printable materials.

Design/methodology/approach – The paper addresses the utilization of polymer nanocomposites as it relates to printable and flexible technology for electronic packaging. Printable technology such as screen-printing, ink-jet printing, and microcontact printing provides a fully additive, non-contacting deposition method that is suitable for flexible production.

Findings – A variety of printable nanomaterials for electronic packaging have been developed. This includes nanocapacitors and resistors as embedded passives, nanolaser materials, optical materials, etc. Materials can provide high-capacitance densities, ranging from 5 to 25 nF/in^2, depending on composition, particle size, and film thickness. The electrical properties of capacitors fabricated from $BaTiO_3$-epoxy nanocomposites showed a stable dielectric constant and low loss over a frequency range from 1 to 1,000 MHz. A variety of printable discrete resistors with different sheet resistances, ranging from ohm to Mohm, processed on large panels (19.5 × 24 inches) have been fabricated. Low-resistivity materials, with volume resistivity in the range of 10^{-4}-10^{-6} ohm cm, depending on composition, particle size, and loading, can be used as conductive joints for high-frequency and high-density interconnect applications. Thermosetting polymers modified with ceramics or organics can produce low k and lower loss dielectrics. Reliability of the materials was ascertained by (Infrared; IR-reflow), thermal cycling, pressure cooker test (PCT) and solder shock testing. The change in capacitance after 3 × IR-reflow and after 1,000 cycles of deep thermal cycling between -55°C and $+125$°C was within 5 per cent. Most of the materials in the test vehicle were stable after IR-reflow, PCT, and solder shock.

Research limitations/implications – The electronic applications of printable, high-performance nanocomposite materials such as adhesives (both conductive and non-conductive), interlayer dielectrics (low-k, low-loss dielectrics), embedded passives (capacitors and resistors), and circuits, etc.. are discussed. Also addressed are investigations of printable optically/magnetically active nanocomposite and polymeric materials for fabrication of devices such as inductors, embedded lasers, and optical interconnects.

Originality/value – A thin film printable technology was developed to manufacture large-area microelectronics with embedded passives, *Z*-interconnects and optical waveguides, etc. The overall approach lends itself to package miniaturization because multiple materials and devices can be printed in the same layer to increase functionality.

Keywords Capacitors, Resistors, Printed circuits, Thin films, Composite materials, Dielectric properties

Paper type Research paper

1. Introduction

There has been increasing interest in the development of printable electronic circuits on flexible substrates to meet the growing demand for low-cost, large-area, flexible, and lightweight devices, such as roll-up displays, e-papers, connectors, and keyboards. Nanomaterials (Friend *et al.*, 1999; Anglos *et al.*, 2004) have attracted a lot of attention for building large-area, mechanically flexible, printable electronic devices. These materials are widely pursued since they offer numerous advantages in terms of ease of processing, good compatibility with a variety of substrates, and great opportunity for structural modifications. In recent years, significant progress has been achieved in the development of semiconductor packaging technology using various printing methods such as screen-printing, ink-jet printing, and microcontact printing. This trend is driven by demand for low-cost, large-area, flexible, and lightweight devices. Since printing is inherently additive in nature, material, and disposal costs are expected to be reduced, resulting in an extremely low net system cost. Most of the research activities in this printable area have been devoted to developing ink-jet solution-processable conductor materials (Blanchet *et al.*, 2003; Huang *et al.*, 2003; Lee *et al.*, 2005; Wu *et al.*, 2006). Printable materials need to be chemically and physically inert to the other functional, dielectric, and photoimageable materials processed in the same layer to preserve the structural and electrical integrity of devices/packages and they have to be operationally stable to sustain long service life. For these purposes, organic and polymeric materials have been widely pursued, since they offer numerous advantages including low-temperature processing, compatibility with organic substrates, stability, and significant opportunity for structural modification. Nanomaterials/composites/hybrids provide the greatest potential benefit for high-density, high-speed, miniaturized advanced packaging. The small dimensions, strength, and the remarkable physical and electrical properties of these structures make them very unique materials with a range of promising applications. Semiconductor devices based on functional polymers, composites, and hybrids are considered to be very promising

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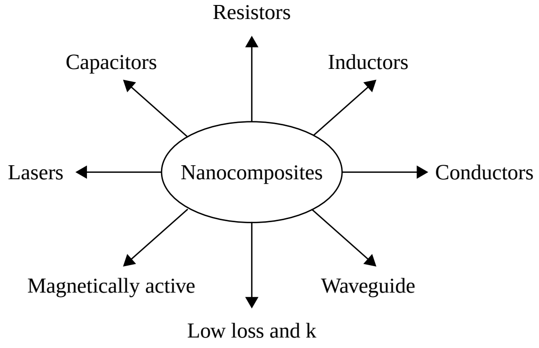
for electronic applications since they may potentially be fabricated entirely using similar printable polymer technologies where different active fillers can be introduced within the same functional polymer system. Several nanocomposites have been reported for advanced packaging applications. Although several nanocomposites used for the advance of semiconductor packaging technology are not always printable, the authors believe that there is potential room for improvement of the existing materials, so that low processing temperature, flexible and cost-effective printable processes, and materials can be developed for large-scale production. An effort in this direction is presented in the paper.

In this work, novel printable materials are reported that have the potential to surpass conventional materials to produce fine structures compatible with organic substrates. Specifically, the electronic applications of printable materials are discussed (Figure 1), such as adhesives (both conductive and non-conductive), interlayer dielectrics (low-k, low-loss dielectrics), embedded passives (capacitors and resistors), and circuits, etc. Printable materials are also being investigated for the fabrication of devices such as inductors, embedded lasers, and optical interconnects. In this work, epoxies have been used as the typical polymer matrix and a range of metal/ceramic fillers with particle sizes ranging from 10 nm to 10 μm. The addition of different fillers into the epoxy matrix controls the overall electrical properties of the composites. For example, the addition of zinc oxide nanoparticles into epoxy shows laser-like behaviour upon optical pumping, and the addition of barium titanate ($BaTiO_3$) nanoparticles results in high capacitance. Thermosetting resins have advantages in terms of manufacturability, processing temperatures, low moisture absorption, high glass transition temperatures (Tg), and versatility make them quite promising for advanced packaging. However, homogeneous dispersions of ceramic particles in the epoxy matrix are a critical step in order to achieve uniform film properties.

2. Experimental procedure

A variety of $BaTiO_3$, ZnO, silver nanoparticles, and their dispersion into epoxy resin were investigated in order to achieve uniform prints. In a typical procedure, ZnO/Ag/$BaTiO_3$ epoxy nanocomposites were prepared by mixing appropriate amounts of the nanopowders and epoxy resin in organic solvents. A thin film of this nanocomposite was then printed on a copper substrate and cured or laminated. The content of the metal/ceramic filler in the composites ranged from 40 to 95 per cent by weight, depending on application. The effects of polymers, surface modifications, particle size, and loading parameters are important for the printing process. Particle sizes below 25 nm are used for ink-jet printing. Titania (~10 nm), silica (~10-20 nm) and silver (15-25 nm) nanoparticles, in pure form or surface modified, can be dispersed in water-based solution. A total of 80 nm or larger particles were utilized in making pastes for screen printing. Dispersion of nanoparticles into the solution is necessary to formulate inks for ink-jet printing. For example, titania and zinc oxide dispersed well in acetic acid-based solutions. Surface-modified nanoparticles typically dispersed well in organic solvents.

Figure 1 Overview of some of the potential applications of nanocomposites in microelectronics

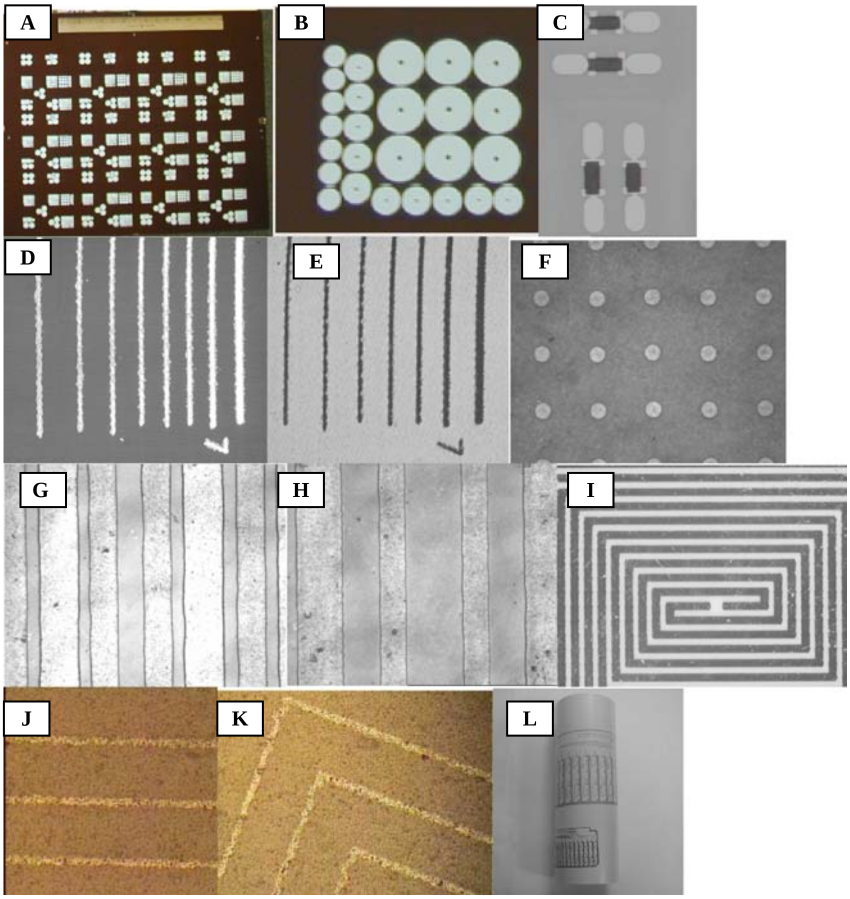


Electrical properties (capacitance, Dk, loss) of the nanocomposite thin films were measured at room temperature using an impedance/gain-phase analyzer (Model 4194 A, HEWLETT-PACKARD). The dielectric constant (Dk)/capacitance as a function of temperature was determined using a precision LCZ meter (Model 4277 A, HEWLETT-PACKARD) at 1 MHz. Surface morphology and particle distributions of nanocomposite films were characterized using a LEO 1550 SEM (scanning electron micrograph). The thicknesses of films were determined by optical microscopy and SEM. An impedance analyzer and a Keithley micro-ohmmeter were used for electrical characterization.

3. Results and discussion

Printable materials have potential applications at all levels of microelectronics (Figure 1). This paper examines the use of nanomaterials in the area of printing technology. Printing processes have several advantages such as selective deposition, repair, and re-print capability. However, printed features with desired properties, thickness, and tolerance present significant challenges. In general, dilute solutions are used for thin ink-jet printing and pastes are used for thick screen and contact printing. Nanomaterial concentration and corresponding viscosity are important for printing processes. Low viscosity is preferable for ink-jet printing processes, i.e. in the range of 7-10 cp. Low viscosity enables the generation of submicron thin structures. Screen and contact printing are better performed using higher viscosity (100,000-150,000 cp) thixotropes and generate 10-25 μm thick features. Conducting polymers, composites, and nanoparticles favour ink-jet printing for transistors and waveguides. Screen/contact printing can be used for making random lasers where surface particles are active. Embedded resistors, capacitors, and conducting circuit lines can use ink-jet or screen printing for different features. Dielectric features are typically large and can use any of the known printing techniques. Figure 2 shows various printings. Figure 2(a)-(e) shows the screen printing processes. Screen print methods can produce line features in the range of 100 μm Figure 2(f)-(l) represent ink-jet printings with minimum line feature size in the range of 30 μm (Figures 2(j)-(k)) and ~50 μm dot patterns (Figure 2). In addition, flexible packages are being developed for a variety of applications. Several classes of flexible materials can be used to form high-performance flexible packaging. Screen and ink-jet printing for low-cost flexible packages are also being investigated. Figure 2(l) represents ink-jet printing on flexible substrates. Figure 2(c) represents screen-printed resistors on

Figure 2 Various printable and flexible materials



Notes: (a) Screen printed capacitors with printed area ((19.5 inch × 24 inch)), (b) enlarged screen print, (c) screen printed resistors, (d)-(e) screen printed ZnO nanocomposites, (f) ink-jet printed 50 μm dots, (g)-(i) ink-jet printed 75-100 μm lines, (j)-(k) ink-jet printed 25-30 μm lines and (l) ink-jet printing on flexible plastics

a PTFE substrate. For selective, large-area deposition, feature sizes, and thickness will dictate printing technology.

3.1 Capacitors and resistors

A novel class of polymer nanocomposites which has shown a high Dk is a $BaTiO_3$ epoxy nanocomposite. These are used to fabricate thin film-embedded capacitors. High temperature/pressure lamination was used to embed capacitors in multilayer printed circuit boards (PCB). The capacitor fabrication is based on a sequential build-up technology employing a first etched Cu electrode. After patterning of the electrode, the nanocomposite can be deposited and laminated within a PCB. Nanocomposites can be directly deposited by printing. Figure 3 shows a flow chart for making screen printed discrete embedded capacitors and resistors. Capacitance values are defined by the feature size, thickness, and Dk of the polymer-ceramic compositions. Figure 4 shows a representative test vehicle (TV) and corresponding cross-sectional views of screen printed embedded capacitors and resistors. Measurement of electrical properties of capacitors fabricated from nanocomposite prints and having areas of $\sim$2-100 mm^2 showed high capacitance density ranging from 5 to 25 nF/in^2, depending on composition, particle size, and thickness of the

Figure 3 Schematic presentation for making screen printable thin film-embedded capacitors and resistors

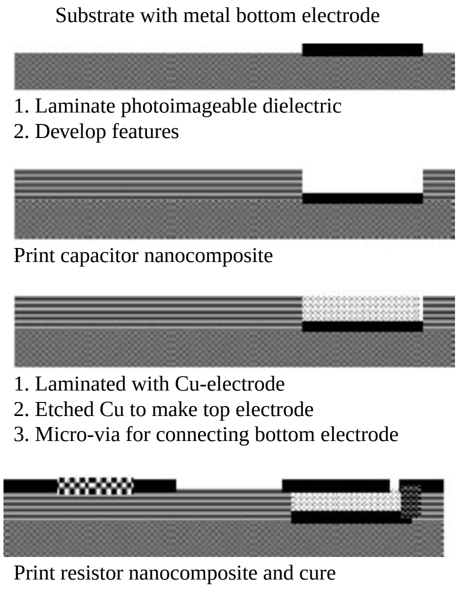
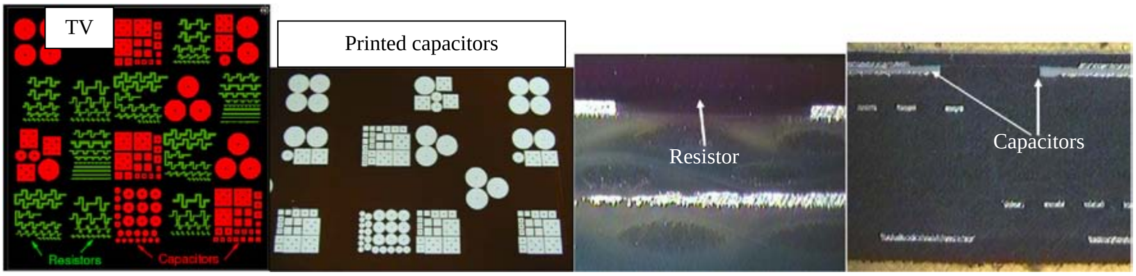


Figure 4 Representative TV and corresponding cross-section view of screen printed embedded capacitors and resistors (Collaboration: Georgia Tech PRC and Nokia)



prints. Thin film capacitors fabricated from 40 to 60 per cent v/v $BaTiO_3$-epoxy nanocomposites showed a stable capacitance density in the range of 5-20 nF/in^2. Capacitors fabricated from a 70 per cent v/v nanocomposite showed a capacitance density of about 25 nF/in^2.

A network analyzer has been used for high-frequency measurements of printable embedded capacitors. The measurements were carried out from 45 MHz to 26 GHz. Figure 5 shows the high-frequency capacitance profiles of 1 cm diameter capacitors. The curve fitting indicates that these capacitors are equivalent to a 1.4 nF bulk capacitance. The excess inductance, including probe, via and embedded capacitors, was estimated to be 60 pH. Figure 6 shows the Dk and dissipation factor measured at 1-1,000 MHz for a $BaTiO_3$ epoxy nanocomposite as a typical representative example. A minimum Dk (3.7) and loss (0.017) were observed for pure epoxy. Addition of high Dk ($\sim$1,200) barium titanate particles into the epoxy matrix increased the overall Dk. The dielectric properties of a nanocomposite are likely influenced in two ways:

1 by microstructure of the composite; and
2 by change in the interfacial or Maxwell's polarization at the interfaces.

For a well-dispersed barium titanate nanocomposite, interface polarization has a great contribution on the dielectric property. According to Maxwell's rule for dielectric mixtures, the measured Dk (composite) values should exceed the corresponding epoxy Dk such that Dk (epoxy) < Dk (composite) < Dk (particle). The dielectric loss increased from 0.01 to 0.025 with increasing frequency.

Figure 5 Impedance profile of capacitors

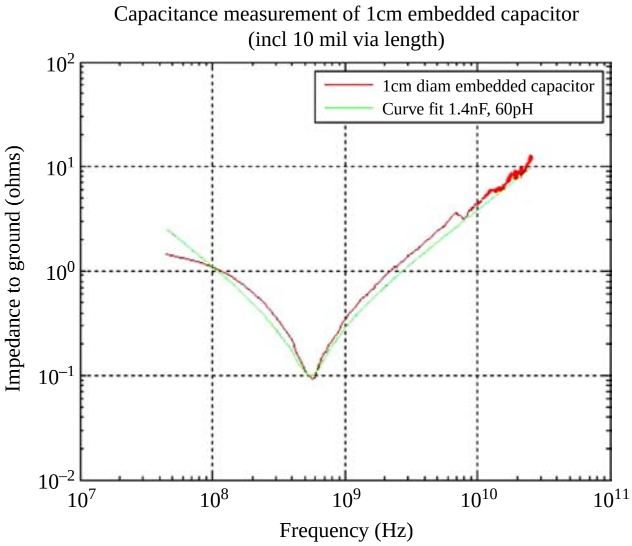
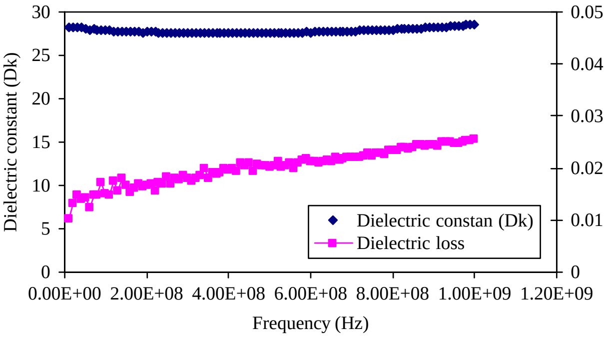


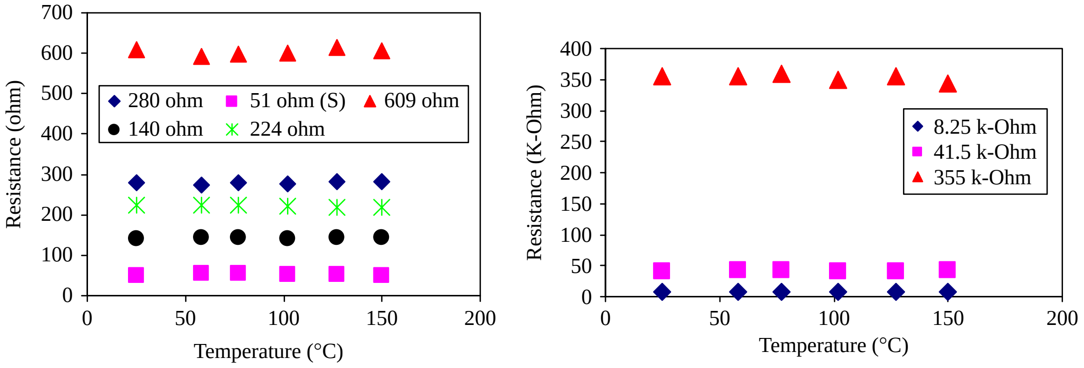
Figure 6 Dielectric constant (Dk) and loss as a function of frequency for printable materials



Nanocomposites are also attractive for resistor applications because variable resistor materials can be formed simply by changing the metal/insulator ratio. These compositions, however, have practical advantage only when they are capable of being printed in the internal layers of circuit boards. Various discrete resistors have been developed with sheet resistances ranging from 1 ohm to 120 Mohm. Resistors in various ranges offer low temperature processing and resistor materials can be printed in the same internal layer. Representative examples of temperature profiles (25°C-150°C) of thin film resistors are shown in Figure 7. The electrical properties of resistors fabricated from epoxy nanocomposites showed a stable resistance over this temperature range.

The reliability of the nanocomposites was ascertained by (Infrared; IR-reflow), thermal cycling, pressure cooker test (PCT), and solder shock. Changes in capacitance after 3 × IR-reflow and after 1,000 cycles of deep thermal cycling (DTC) between -55°C and 125°C were within 5 per cent. Changes in capacitance after IR reflow (assembly), pre-conditioning (3 ×, 245°C), and thermal shock up to 1,400 cycles (-55°C – 125°C), for large, medium, and small embedded capacitors, were <5 per cent. Most of the nanocomposites in the TV were stable after IR-reflow, PCT, and solder shock. The change in conductivity of electrically conducting adhesives after 3 × -IR reflow at 220°C was < 5 per cent. Some of the low-loss materials were also exposed to PCT (four hours) followed by a 15 s solder dip at 260°C. PCT and solder shock sometimes

Figure 7 Change in resistance with temperature



caused delamination. In general, solder dip/shock picked up the PCT-induced defects and caused delamination. Initial PCT and solder dip experiments did not show any delamination. Table I summarizes the reliability of printable embedded capacitors.

3.2 Inductors

Ink-jet printing of spiral structures can be used to form inductors. The spacing in the spiral and the resistance will dictate the quality of the inductors. High resistance causes thermal loss and, therefore, is not suitable for inductors. In this work, multi-metal layers have been deposited on ink-jet printed lines to increase current carrying capacity or conductance. High-conductance spirals can generate higher magnetic fields at the same voltage and thus can provide higher inductance in smaller packages. A variety of multi metal layers have been used including electroless copper, immersion gold, electroless gold, electroless palladium, electroless nickel, etc. Figure 2(i) shows a representative example of spiral inductors. Multi-metal layer deposition on spirals reduces the line resistance to hundreds of milliohms.

3.3 Conducting adhesives for interconnects

Low-resistivity nanocomposites with volume resistivities in the range of 10^{-4}-10^{-6} ohm cm, depending on composition, particle, size, and loading, can be used as conductive joints for high-frequency and high-density interconnect applications. Metal-to-metal bonding between conductive fillers provides electrical conductivity, whereas a polymer resin provides better processability and mechanical robustness. Materials can be printed or filled in a joining core to fabricate *Z*-axis interconnections in laminates. Conductive joints were formed during composite lamination using an electrically conductive adhesive. The adhesive-filled joining cores were laminated with circuitized subcomposites to produce a composite structure. High-temperature/pressure lamination was used to cure the adhesive in the composite and provide interconnection among the circuitized subcomposites. Figure 8 shows an optical photograph of a cross-section of a PCB constructed using glass-cloth-reinforced dielectric materials. The construction was assembled from multiple multi-layer sub-composites. The inset shows a somewhat enlarged view of a land-to-land adhesive connection.

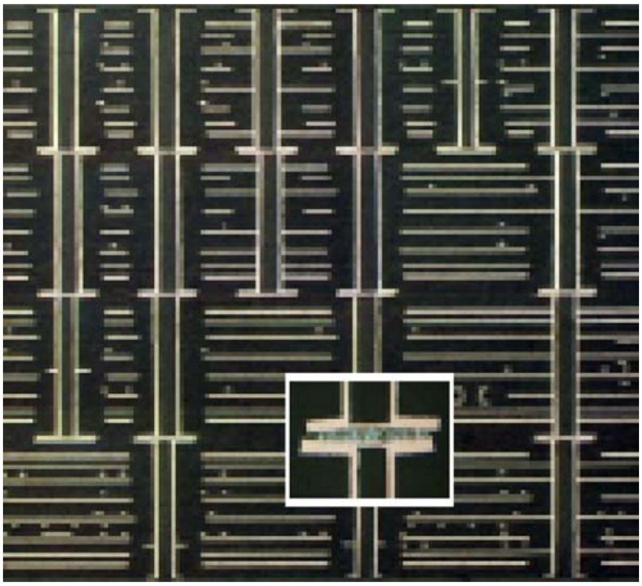
3.4 Printable ZnO

ZnO has been proposed as an interesting material for optical devices in the blue to ultraviolet (UV) wavelength region because of its large direct bandgap of 3.4 eV. ZnO-based semiconductors can cover nearly the same wavelength range as GaN. The excitonic binding energy of ZnO is much larger than GaN-based compounds. Much attention has been given to ZnO-scattered systems that, upon pumping, exhibit laser-like emission described by the term random laser. A number of ZnO-based random lasers, including ZnO polycrystalline film, powders (Ozerov *et al.*, 2004), ZnO microlasers (Ling *et al.*, 2001), ZnO-based hybrids (van Soest *et al.*, 1999; Bahoura *et al.*, 2002), etc. have been developed. In conventional lasers, photons reflected back and forth through a cavity stimulate the emission of more photons, thereby helping to build up an intense coherent radiation beam (Wiersma, 2000). A similar effect can be produced in a disordered medium containing semiconductor particles or in a finely ground semiconductor powder. If the particles or grains are close enough – less than the wavelength of light – the photons form closed loops. As a result, the light is scattered passing through the same grains, just as in an ordinary laser, light bounces back, and forth between the mirrors leading to light amplification. Wiersma (2000) suggested several possible applications of ZnO-based random lasers in a variety of new miniaturized optical devices. Das and

Table I Environmental test results

Test	Property	Results
Solder shock (15 s dip at 260°C)	Bonding	Passed
PCT + solder shock	Bonding	Passed
IR-reflow (245°C, 3 ×)	Capacitance	< 5%
Bake (150°C /four hours) + 1,000 deep thermal cycle (− 55°C − 125°C)	Capacitance	< 5%
IR-reflow + 1,400 DTC	Capacitance	< 5%

Figure 8 Photograph of *z*-interconnect laminate shown in cross section



Giannelis developed a variety of ZnO polymer nanocomposites. Epoxy, polydimethylsiloxane, and polymethylmethacrylate (PMMA)-based ZnO nanocomposites show lasing at around 385 nm (blue-violet region). When ZnO is dispersed in a fluorescent polymer like poly[2-methoxy-5-(2′-ethylhexyloxy)-*p*-phenylene vinylene], it shows lasing at around 610 nm (red region). Furthermore, zinc oxide is useful as a piezoelectric and in sensor materials. This material can be used as filler for capacitance layers where ZnO improves the microstructure and film quality of barium titanate epoxy capacitors. Printable ZnO has been developed for a variety of fine structures and it is possible to print different line widths and spacing ranging from about 3.5 to 10 mils. Smaller features, such as ~2 mil dots, can also be printed. All these features can be used as random lasers, or capacitance, or optical shielding layers. Thus, it is possible to use printable ZnO as a multifunctional material for devices.

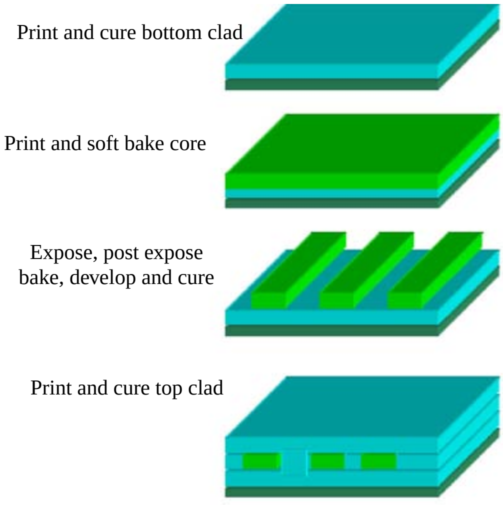
3.5 Magnetically active nanocomposites

Another attractive nanocomposite is a magnetically active nanocomposite. Magnetic nanoparticles have excellent microwave absorption properties. These are widely used as electromagnetic absorbers in radiation shielding. Also, high-density recording media uses these particles (Vian *et al.*, 1996; Yamamoto *et al.*, 1997). These materials are also used in microwave-based communication systems functioning at frequencies of the order of 10^9 Hz due to their low crystalline anisotropy (Gotic *et al.*, 1998; Pannaparayil *et al.*, 1988). Preparation of nanocrystalline ferrite materials has been described in our previous paper (Das, 2001). Magnetically active nanocomposites can function as passive magnetic devices and the nanocomposites are used to control inductance of the circuitry. A great deal of activity has been directed towards the development of printable magnetic nanocomposites. In a typical procedure, different magnetic nanoparticles can be embedded into an epoxy matrix to provide passive magnetic devices such as inductors, antennas, and transformers.

3.6 Optical waveguides

Waveguides are important for high-speed applications. Several polymer and nanocomposites are reported to be useful as waveguides. Zhang *et al.* (2006) reported silver nanoparticle and rhodamine B-based planar polymer multi-model waveguides. The silver nanoparticle concentration enhances the optical properties (fluorescence) of rhodamine-doped PMMA planar waveguides. Saj *et al.* (2006) that described plasmon waveguides are composed of silver nanoplates arranged in several geometries to find the one with the lowest attenuation. They have investigated 500 nm wavelength light propagation along different chains of silver nanoplates of sub-wavelength length and width and wavelength-size height. Yeo and shin *et al.* (2006) developed a new polymer-silica hybrid thermo-optic switch with significantly reduced cross talk. The top cladding and the core layers are composed of polymer, while the bottom cladding layer is made of silica. Among various techniques, UV curing, micro-moulding, and replication are successful processes to fabricate polymer waveguides. The polymer waveguides depicted in Figure 9(b) were fabricated using an ink-jet process-compatible optical polymer. Owing to the spatial resolution and drop-size limitations of current ink-jet equipment, waveguide channels cannot be "inkjet printed". Instead, high-resolution lithographic processes must be used to define the actual waveguide channels. Although screening of the waveguide materials is possible, ink-jet processes provide excellent material deposition and coating, both in terms of flexibility, with accurate and uniform thickness control in the range of two to 3μm Figure 3(b) shows the ink-jet printable, large-area, selective deposition of waveguide materials. The desired thickness of cladding or core layer can be achieved by single or multiple printing passes. Contact and non-contact photolithographic processes have been used with high-quality photomasks for defining waveguide channels.

Figure 9 Schematic presentation for making waveguides



(a)

(b)

Notes: (a) Printable; (b) optical

Figure 9(a) shows a flow chart for making optical waveguides. Good-quality waveguides with low loss (approximately 0.05 dB/cm) at 850 nm can be fabricated using ink-jet printing. The major challenge of deploying this technology is in the formulation of the optical polymer with suitable viscosity and adhesion properties.

3.7 Low-k and low loss composites

Low-loss materials are important for high-frequency and high speed applications. Low-k materials are useful to reduce the dielectric thickness of the resulting circuit substrate. The rapidly growing wireless industry requires high-performance materials to build low-loss, high-density, thermally stable integrated packages. GHz operating frequency systems require substrate materials with lower loss (Df), low Dk, and good power handling characteristics, which are important in many of these applications. Low loss is a critical requirement for lightweight portable devices for long battery life. Low-k dielectrics not only lower line-to-line capacitance, but also reduce cross talk problems between traces. Organic polymers such as divinyl siloxane benzocyclobutene, a silicon-based polymer with high organic content, and poly(arylene)ethers are some examples of low-k materials. Fluoropolymers, fluorinated polyimides, polyimide-silica hybrid, and bismaleimide-triazine in combination with epoxies have been used as low-loss and low-k dielectric materials. This paper describes filled printable low-loss and low-k materials compatible with laminated organic substrates. Ceramic or organic filled polymer systems were introduced where ceramic/organic fillers and content dictated the properties of the composites. Pure silica and multi-component silica, boron nitride, alumina, and several other low-k and loss fillers were used as printable composites. Figure 10 shows the variation of Dk and loss with frequency. Dk decreases with increasing frequency. Screen printing and dispensing techniques are generally used for printing dielectric materials.

4. Conclusions

Printable materials are promising, not only because they are versatile but also economical compared to other methods. A variety of materials suitable in printable processes for the fabrication of selective and localized embedded components in printed wiring board/laminate chip carrier has been developed (Figure 11). The materials and processes enable fine feature sizes and controlled thickness of deposited layers. This result is accomplished by ink-jet, screening, and contact printing and dispensing processes. Experiments demonstrated that ink-jet printing and subsequent metal layer deposition are suitable for inductors, whereas screen or contact printing is suitable for conducting adhesives for interconnect applications. Capacitors, resistors, ZnO, and waveguide materials can use either ink-jet or screen/contact print processes based on their requirements and viscosity of solutions. Capacitors fabricated using a printing process showed high capacitance and low loss and are reliable after IR-reflow and DTC. Nanomaterials can produce variable resistances ranging from ohms to Mohms. Low-k and low-loss materials can also be fabricated from nanocomposites. Overall, printable materials will be useful to produce multi-functional complex electronic packaging. The results also suggest that printable nanomaterials may be attractive for the roll-to-roll manufacturing of large-area microelectronics such as roll-up displays, e-papers, keyboards, radiofrequency structures, transistors, photovoltaics, medical devices, etc.

Figure 10 Dielectric constant and loss as a function of frequency for an epoxy-filled system

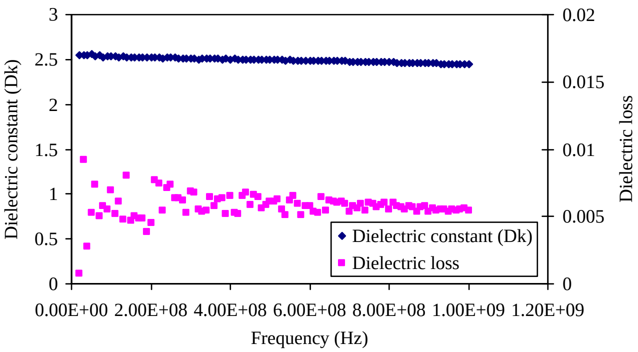
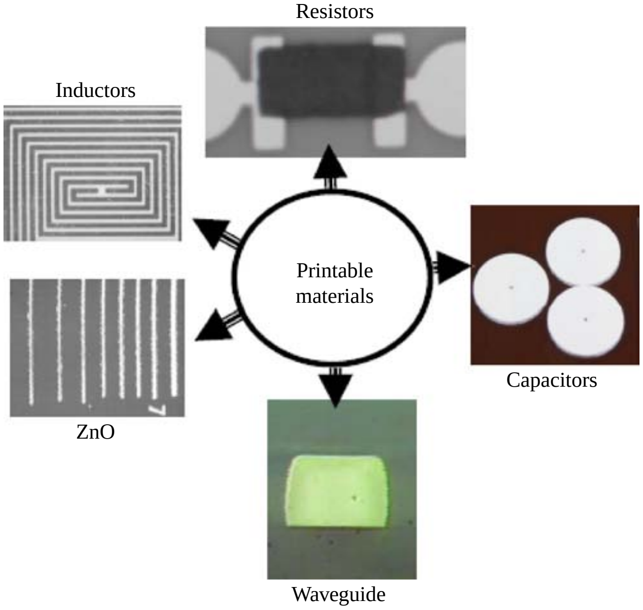


Figure 11 Printable materials in microelectronics. Multiple materials can be printed in the same layer



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