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# Development of a multi-material additive manufacturing process for electronic devices

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**Abstract**

In order to increase the versatility of additive manufacturing multimaterial processes, a hybrid system has been developed, which is capable of combining 3D printing technology by DLP (Digital Light Processing) with a two-dimensional Drop-on-Demand Inkjet printing system.
Through DLP technology based on digital micromirror devices (DMDs) it is possible to build up 3D geometries layer-by-layer using polymerization of photosensitive resins. Concurrently, while the construction process is performed, the InkJet printing system is used to deposit tiny drops of conductive inks on the substrate generated, which will thus constitute an electric circuit embedded within the three dimensional structure.
On the other hand, photosensitive resins have been filled with Low Temperature Co-firing Ceramic (LTCC) particles, in order to modify the basis properties of the part by using sinterizable slurries. Finally the challenges in the sintering process for achieving functional parts are discussed and a few prototypes have been built in order to validate this technology.


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## 1. Introduction

Printing processes have traditionally been described as methods of reproducing texts or images on paper. Nowadays the continuous evolution of these technologies has rendered such definition obsolete by referring to it as a conventional print. The functional printing arises as an umbrella term for all types of printing. Under this concept, the functionality of the reproduced element precedes the reproduction action; terms like printing electronics, 3D-printing, bioprinting or additive manufacturing are all concepts encompassed within functional printing term.

3D-printing is the main representative of functional printing that has been available during more than 30 years since the patent which described the stereolithography (SLA) was registered in 1984. The additive manufacturing (AM) on the other hand, is a more recent term. In both cases, the description of a collection of techniques that allows reproducing 3D-objects from a digital file by the sequential addition of energy and/or material is performed. At the beginning, these different techniques allowed the reproduction of objects using a single material where the piece is materialized layer-by-layer offering the possibility of overcoming geometric limitations imposed by other more traditional techniques. Year by year, technological evolution has allowed reaching high levels of detail and perfection. Recently, following the high expectations generated by these reproduction systems, these technologies have been immersed in the requirement to equip these systems with the possibility of printing with multiple colours and materials. The latter has been one of the limiting factors of these technologies.

At the same time to the development of these three-dimensional reproduction systems, conventional printing technologies based on InkJet printing systems have also undergone a huge evolution. The development of electronics and its miniaturization allowed the development of drop-on-demand (DOD) systems. And with it, those powered by piezoelectric the limitations on the printed matter have been significant reduced. Nowadays InkJet printing is presented as one of the most promising digital printing techniques. The printed fluid (ink) pays a great role; the ink determines the functionality of the reproduced element. In this regard, printed electronics refers to the printing of conductive or electroluminescent or photovoltaic inks.

It is expected that printed electronics will make low performance; low cost generalized electronics much easier and accessible, establishing itself as a reality available for consumption.

This work seeks within the functional printing umbrella to combine three-dimensional printing with printed electronics into a single multimaterial hybrid printing system in a simple way and thereby provide the reproduced objects with new functionalities, with the aim of increasing the added value to the customizable printed parts, thus justifying the high cost of additively manufactured objects.

## 2. Digital Light Processing technology

The digital light processing (DLP) technology is the younger sister of SLA systems. In this technology the provided energy is irradiated by layer and not by point; the material is also added layer-by-layer offering a greater speed of construction than its elder homologue, especially in the reproduction of complex shapes or layers with big surfaces. Taking advantage of the exceptional spatial control and versatility of photopolymerisation reactions, such technology can offer a highly printing resolution in comparison with other technics. The latest developments on digital devices such as light crystal displays (LCDs) and digital micromirror devices (DMDs) presents at relatively low costs powerful tools that can simultaneously and dynamically control energy inputs of a projection imagen.

As a first step, a 3D CAD model of an object to be reproduced is sliced by a set of horizontal planes. Then, each slice is converted into a two-dimensional mask image. The related mask images are sent to a DMD and projected onto the resin surface to be selectively cured to form a layer of the object. By repeating this process, 3D objects can be formed layer-by-layer on a platform.

### *2.1. Photosensitive resins*

In a photopolymerization reaction, a liquid resin is hardened when is exposed to an energy source such as ultraviolet (UV) or visible light. There are two types of reaction mechanisms: free radical mechanism usually through double or triple bonds of (meth)acrylate - and cationic mechanism - through epoxy groups.

Typically, the photosensitive resin contains monomers, a photoinitiatior and others additives to tune its properties, as well as reaction inhibitors or accelerants agents. The photopolymerization phenomena are mainly explained by three basic steps: initiation, propagation, and termination.

The initiation process occurs when the resin is exposed to light. In this step the photoinitiators absorb the photons of a certain wavelength, generating highly reactive free radicals. During the propagation process, the radicals previously generated activate de polymerization by reacting with double and triple bonds of the monomer. This results in a radical monomer which grows by successive reactions with other monomer molecules. Finally, the termination process happens when the end of one radical monomer chain links with another.

The optimal printing parameter is strongly dependent on the resin formulation. One of the most important parameters is the critical energy dose, which is defined as the minimum energy per area that allows the photopolymerization reaction. The cured depth, another critical parameter, is defined as the thickness of the cured resin. This parameter depends on the energy intensity, exposure time (both related with the energy dose), amount of inhibiter agents, monomer nature and the amount of photoinitiator.

Apart from the resin properties, there are other parameters that affect the printing process such as the resolution of the light projector. It is known that the material´s refraction index of the projector lens system depends on the wavelength, therefore the focal distance will differ depending on the projected wavelength. Consequently, the different refraction indexes of a polychromatic source light can induce defocus effects on the printed part. Moreover, by fitting the absorbance peak of the chosen resin with the wavelength of the monochromatic light, one significantly improves the features of the printing process. In summary, the wavelength of the projected light must be as closest as possible to the wavelength associated with the maximum absorbance of the photoinitiator in order to effectively maximize effective its excitement.

### *2.2. Photosensitive ceramic slurry*

Additive manufacturing techniques have been successfully used in the reproduction of prototypes whose complex geometries fulfill a structural function, although often the limitations imposed by the printable materials, restricted to photosensitive resins, reduces their applications, in particular in final printed parts. In this sense, the use of these technologies to manufacture ceramic materials can result in a big advantage.

However, some limitations have to be overcome. Resins physical properties (mechanical, thermal, electrical, and magnetic) can be improved by filling the photopolymeric resin with ceramic particles. The main principle of these resins is the same as explained previously, but in this case, the polymeric material is also used as a matrix to trap the ceramic particles. The addition of particle fillers into the resin will modify its characteristics, such as its rheological behavior, photosensitivity and scattering of light, which are directly related with its printing capacities. In general terms, the presence of ceramic fillers dramatically increases the viscosity and decreases the curing sensitivity (consequently lowering the cured depth).

Whenever, ceramic materials are used as a final product and not as a composite (ceramic particles and polymeric matrix). The final properties must be achieved by the properly removing of the polymer in a debinding process and posterior sinterization. Therefore, the main challenge of additive manufacturing of ceramic materials is to control these features accurately, which will in turn enhance the printability and applicability of those outstanding materials.

## 3. InkJet piezoelectric DOD technology

InkJet technology is a non-impact dot-matrix printing system in which droplets of ink are jetted from a small orifice directly to a specified position on a substrate to reproduce the desired patterns. Those patterns to our eyes can become a point a line, letters or images. On the other hand, the ejected droplets can be generated by two different mechanisms: continuous ink-jet (CIJ) printing or drop-on-demand (DOD).

In spite of CIJ printing operates at much-faster droplet-generation rates than DOD printers, the need to use an electrically conducting fluid and the possibility of its contamination during the recirculation process limits its applications. Hence, for this study, we have confined our attention to DOD printing, where the droplets are only ejected when required and the printhead is mechanically positioning above of a desired location over the substrate.

Since its invention by Canon and Hewlett-Packard in the end of the 70`s this technology has evolved in parallel with the progress of the computer itself. The thermal InkJet technology (TIJ) reduced the costs per nozzle and overcame the bottleneck of the miniaturization. The reduction of the printhead cost allowed the replacement of it every time the ink deposit was empty. The concept of a disposable InkJet cartridge helped to overcome its problems of reliability and since then this printing system has had a resounding success to the point that there is an ink printer beside to almost every computer in many homes.

More recently, the applications of InkJet technology has been extensively expanded. Replacing the colour inks by functionalized inks with a specific purpose and adapted to the conditions for its ejectability, new applications were discovered for these deposition technologies. However, TIJ technology, since it is based on the thermal action of the resistance, limits the usable materials which gave rise to the resurgence of piezoelectric InkJet technology (PIJ), which today dominates the market of printing micromanufacture.

In the piezoelectric InkJet printing, depending on the piezoceramic deformation mode, the technology can be classified into four main types: squeeze, bend, push, and shear [1]. In this study, a cartridge of bend type from Fujifilm has been used. Particularly the model Dimatix DMC-11610 (Fig. 1.a) able to draw droplets of 10 picoliters by its 16 nozzles spaced at 254 µm [2].

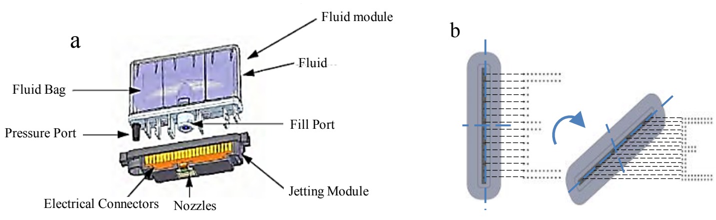


Fig. 1. (a) Schematic representation of the FUJIFILM Dimatix (DMC-11610) Cartridge used a) [3]; (b) Schematic representation of the printing process and the adjusting of the resolution by the rotation of the printhead.

## 4. Hybrid 3D printing

The integration of multiple printing technologies in a unique system has been carried out for some time, in 2005 F. Medina et al. [4] carried out the combination of a SLA system with a direct writing system (DW) in an hybrid printing system. Given the enormous potential of the manufacture of complex geometries with embedded circuitry, since then there has been a considerable interest in this field. More recently, advances in ink dispensing methods have allowed this technology to be more attractive for the manufacturing of complex 3D systems. 3D-objects with a relatively high density of passive components integration (resistors, coils and capacitors), in which the placement of active electronic components (operational amplifiers, transistors, etc.) is now possible, providing novel properties to the reproduced objects and also the ability to interact with its users at a higher level [5,6].

The system presented here represents a first step in the development of multi-material hybrid systems in which bimaterial pieces are reproduced. This system opens the possibility of combining greater number of materials through the use of multiple deposits of resins and inks. In this regard, Maruo et al. [7] was the first who presented a multi-material SLA system through the manual exchange of vats, Wicker et al. [8] extended Maruro's work by developing a multiple vat carousel system by automating the construction process including the cycle of washing, curing and drying between building materials. Regarding systems where the contribution of energy is made by layer, Zhou et al. [9] presented a similar bimaterial printing system using DLP technology and adding a step of cleaning and drying to the carrousel.

Concerning InkJet system, the implementation of multiple cartridge or even printheads in the printing carriage would open the possibility of combining multiple materials and in different graduations. On the other hand, being a printing system without impact, the deposition of these materials can be carried out on any flat substrate being able to be this one created additively in an earlier stage as is our case. The combination of multiple materials in both, a carousel of deposits and multiples cartridges or printheads could provide great versatility to this hybrid equipment.

### 4.1. Hybrid DLP/InkJet machine setup

The hybrid printing system presented here combines a three-dimensional DLP printing with an InkJet DOD ink deposition system actuated by piezoelectrics. Both systems offer high speed and precision at low cost and the integration of the InkJet system has been carried out thanks to a top-down configuration of the DLP system.

#### 4.1.1 Projector, XY and Z resolution

In DLP printing the XY and Z-resolution are different. The XY-resolution is determined by various parameters: the resolution of the DMD, lens distortions, the distance of the projector to the free surface of the liquid resin and the scattering generated by blockers and shrink. The theoretical resolution achieved with a full-HD projector with a luminous power of 3000 lm on a flat surface with a width of 122.5 mm, results in a maximum XY-resolution of 63.8 µm/pixel. The Z-resolution is determined by the mechanical resolution and is also influenced by shrink.

Since stepper motors are been used to drive the spindle where the build platform is attached to, the positions of that platform are discrete, and due to the minimum stepping angle and the lead of the screw of spindle the mechanical resolution can be determined. Thus, theoretical achievable Z-resolution is 12.5 $\mu m$. However, the precision of the spindle and consequently in the platform could be slightly modified by the backlash of the couplings.

Due to being a top-down configuration system, the platform is immersed in the resin reservoir during the printing process of the 3D-structure, consequently the level of resin in the reservoir has to be controlled. For this reason a pumping system has been incorporated. Also, with the objective to remove the possible bubbles and irregularities produced in the free surface of the deposit and through the solidification of the layers do not transmit them to the reproduced object, a wipper has been added to the system (Fig. 2). This device also has as a function to pour the excess of resin to the channel that surrounds the deposit generated as a consequence of its own surface tension.

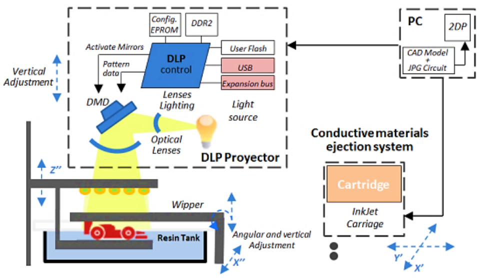


Fig. 2. Illustration of the DLP system in which the 3D structures are performed and the Inkjet system were the circuit are reproduced.

#### 4.1.2 InkJet piezoelectric DOD system

The printhead allows the exchange of cartridges and in addition to the translation movements, also offers the possibility of a slight rotation of the cartridge in order to reduce the vertical drop spacing, modifying the resolution capacity of the system (see Fig. 1.b). On the other hand, the ejection control of the inks through the cartridge is carried out by means of the combination of three signals: one of temperature control (resistance/thermistor), another of excitation in order to keep the meniscus vibrating and thereby avoiding nozzle obturation and finally, a third signal of excitation of the piezoelectrics actuators responsible for the ejection of the droplets where its shape and duration are conditioned to the rheology of the ink.

The process of deposition of the ink through the InkJet system can be splitted in four steps: firstly, the generation of the droplets through each nozzle of the cartridge and therefore its ejection, it has to be coordinated with the lineal displacement of the carriage. Secondly, after the ejection the droplets that describe a parabolic trajectory decelerated by the air friction are reshaped into a spherical form. Thirdly, once the drops impact at the desired location on the substrate, a sequence of events takes place, where after a dynamic transition where the oscillations are damping, the capillary-driven flow occurs [10]. Finally, as a fourth step, the fixation of the droplet onto the substrate takes place;

then, a phase change of the ink occurs, the pass from liquid to solid is needed. This process can be driven either by ink solidification and evaporation of the solvents or chemical reaction of a polymer, depending on the ink nature.

The control of those previous steps allows the reproduction of the digital files on a substrate. Conventional graphics printing requires patterns made-up of isolated drops to produce a pixilated image. In our case, functional printing require the overlapping of the droplets in order to form electrically continuous features. Thus, the behaviour and the interaction of the droplets, more or less scattered, deposited onto the substrate conditioned the stability of liquid beads or lines or more complexes 2-D patterns. 3-D structures are produced by sequenced layers overprinting, but in this case the droplets interact with a solidified former deposit which performs the substrate functions.

### *4.2. Integration of both systems*

In order to prevent that the photosensitive materials could fouling and hampering the mechanical systems, they have been arranged remotely and protected from them. On the other hand, in order to avoid that the different mobile elements of the hybrid equipment could hinder or even worse interfere with each other; it has had to devise a geometric configuration that allows the integration of them. For this purpose it has been considered to locate these distributed in two parallel planes vertically separated. In this sense, the vertically moving construction platform serves as a link between both printing planes allowing the integration of DLP and InkJet systems into single device.

Nevertheless the reproduction of the object is carried out by the alternation of both processes with a view to print a kind of composite made by dielectric 3-D structure and conductive tracks deposited over the last photopolimerized layer to finally obtain combined circuitries architectures embedded within the tridimensional structure.

## 5. Results and demonstrators

With the aim of demonstrate the feasibility of the hybrid printing of the system discussed above, some of the tests carried out are presented below.

### *5.1. DLP green parts and sintering process*

Photosensitive slurry based on ceramic particles and aliphatic acrylates monomer was optimized considering the rheological requirements imposed by the developed DLP system. The ceramic particles used on the formulation is a low temperature co-fired ceramic (LTCC, 51528 B Glass Powder, HERAEUS) with an average particle size, D50, around 3.4 μm. The homogenization of the slurry was achieved using planetary ball mill working at 300 rpm. To increase both the homogeneity and the percentage of ceramic particles without agglomerations, an anionic dispersant was added and accordingly optimized. The dispersant concentration was adjusted for the lowest viscosity of the suspension, thus ensuring optimal particle dispersion. The rheological measurements were carried out on a TA Instruments Discovery HR-1 rheometer with a 1º cone plate geometry at strain rates of 2–200 $s^{-1}$. The resulting formulation contains 67 wt.% of ceramic particles and 1.56 wt.% of dispersant with respect to the powder content, which results in a non-Newtonian pseudoplastic behavior at room temperature, with a viscosity of 2.48 Pa·s at a shear rate of 2 $s^{-1}$.

The DLP process was validated with the fabrication of a cubic and a cylindrical geometry, as it is shown in Fig. 3. The dimensions were 20.25x15.85x1.9 mm and 20.40x15.60x3.6 mm of high, width, and thickness respectively, for cubic (Fig. 3.a) and cylindrical shapes (Fig. 3.b). The printing was performed with 50 μm of layer thickness at full intensity of visible white light with 4 s of exposure time. However Fig. 3.a. also shows some superficial printing defects that can be optimized by adjusting the wipper height system allowing a better homogeneity of the deposited.

To obtain the final ceramic part, the debinding and sintering process must be performed. The debinding process was achieved from 24°C up to 240°C and from 240°C up to 460°C, both temperatures with a dwell time of 30 minutes. It is known that the formation of gases at high temperature results in internal stresses; hence a low temperature rate was used to adequately decompose the polymeric material, thus avoiding the above mentioned issue. After the debinding process, the part is free of any organic material, thus the sintering process can be properly performed. The sintering process was raised up to 870°C, leading to a final ceramic part with shrinkage of 20%.

As it can be observed in Fig. 4, the sintered part preserves the green part geometry without cracks, meaning that both debinding and sintering processes were accurately performed. The scanning electron microscope (SEM) image (Fig. 4.c) shows a crack-free surface and a homogeneous distribution of the grain particles in the glass matrix, indicating a good dispersion of the initial slurry.


Fig. 3. Green printed parts of the photosensible slurry of LTCC. Front, top and tridimensional view of them; (a) cubic geometry; (b) cylindrical; (c) SEM magnified view at x1.5k of the cubic geometry.

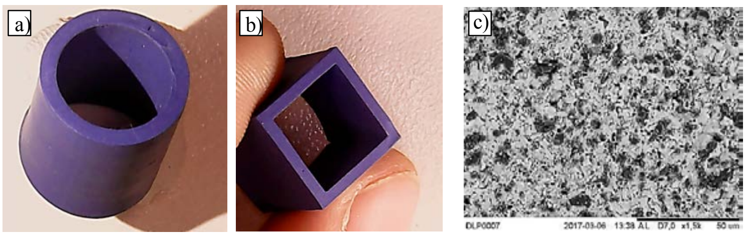


Fig. 4. Sintered printed part made with photosensitive slurry of LTCC. (a) Three-dimensional view of the cylindrical geometry; (b) cubic; (c) SEM magnified view at x1.5k of the cubic geometry.

### 5.2. InkJet 2D patterns

Parallel to the DLP tests, InkJet deposition and its analysis has been also carried out. In this regard, by adjusting the piezoelectric excitation signals of the cartridge as a function of ink rheology and the drop spacing as a function of the affinity of the latter with the various substrates (glass, photosensitive resin or LTCC slurry), two-dimensional electrically continuous patterns have been reproduced. The fixation of stable beads formed by droplets has been carried out by UV-A radiation. As ejected material, a nanoparticle-based silver ink (ANP silverjet DGP HRA) supplied by the company Advanced Nano Products Co., Ltd has been used.

The deposition process has been completed through the displacement of the printhead, which is moved horizontally (X axis) through a belt system driven by a reduction motor where each step of this equals a jump of 20 µm. The printing plane (XY) is completed thanks to the uncoordinated discrete displacement in the direction of the Y axis by means of the spindle and its coupling with almost zero backlash, where each step of the motors equals a jump of 12.5 µm. Fig. 5.a. shows the digital circuity pattern while Fig. 5.b. and 5.c. show the silver InkJet reproduction onto a porous paper and a detailed magnified image of the test respectively.

Fig. 5.d.e.f. shows the test circuit reproduced on different substrates before and after curing of the ink, it should be said that this circuit has dimensions of 14.30x9 mm and with conductive tracks 200 µm wide. On the other hand, in the images of Fig. 5.f, it can be appreciated the reproduction of the above circuit on LTCC substrate in green, where after curing the ink at 170°C for 20 min and after its sintering in three stages, it is shown in detail that the electrically conductive tracks resist the thermal process (resistivity ~$10^{-6}$ Ω·m).

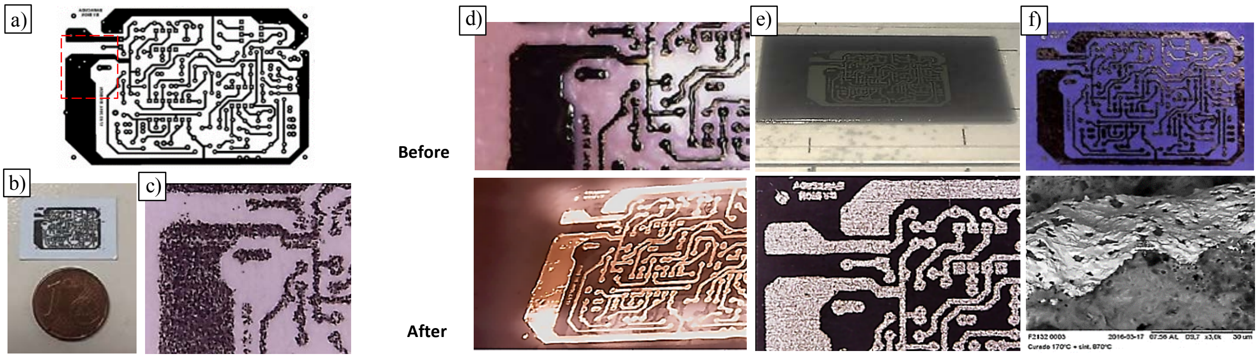


Fig. 5. (a) Completely image of the test circuit; (b) Its reproduction on paper; (c) a detailed magnified view of it. Reproduction of the circuit on a more hydrophobic substrate before and after curing: (d) glass sheet; (e) photosensitive resin (Fun-to-do DB); (f) slurry in LTCC.

## 6. Conclusions and future work

Various tests carried out with the hybrid multi-material equipment developed and described before show that it is feasible to combine in a single device three-dimensional reproduction through a DLP 3D-printer with a deposition InkJet system of functional materials.

However, the use of photosensitive ceramic slurries poses certain challenges requiring further investigation, nevertheless a ceramic part was successfully achieved with minor printing defects.

To date, conductive tracks of 200 microns have been achieved on substrates previously photopolymerized with layers between 25-50 µm thick and it has been verified the possibility of connecting two conductive tracks vertically separated by a slim photopolymerized layer between them.

Further research will analyze the influence of dimensional constraint on printed circuits and it will seek to generate interconnected multilayer circuits immersed in the three-dimensional structure. Finally with the use of surface-mount device chips will seek to provide new properties to reproduced parts and a possible link it to the digital world, adding greater added value to the fully customized ceramic parts printed.

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