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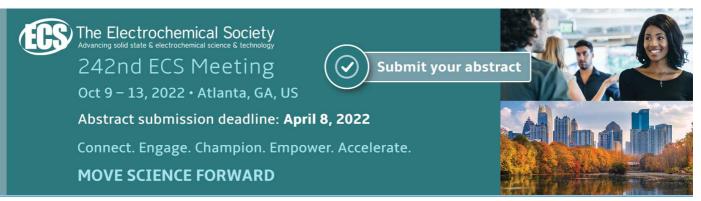
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Research on desktop 3D Printing Multi-Material New Concepts

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Abstract: 3D printing or Additive Manufacturing (AM) was originally born as a mono-material technology. And, nowadays, most of the applications are still using only one material. AM has a lot of potential but has not yet been fully explored, and access to the creation of multi-material products is an example of it. One of the most interesting areas is the introduction in the same part of materials with different rigidities, stiffer and softer areas, with differentiated values of mechanical strength and viscoelasticity. In the present work, a general vision of Additive Manufacturing under the vision of mono- and multi-material processes is given, and some existing 3D printing multi-material experiences related to Material Jetting (MJ) and Material Extrusion (ME) are briefly described. But it is in this latter field, linked to Desktop 3D printing (more accessible than typical proprietary industrial equipment), where on-going research could easily be spread: five research ME concepts are then presented, from a revolver print-head to silicone UV 3D printing, with their initial embodiment in the form of prototypes or/and testing, as a way to verify the difficulties that would be encountered in the transition from research to reality.

Keywords: Multi-material, Additive manufacturing, 3D printing, Material Extrusion.

1. 3D Printing, Originally a Mono-Material Technology

3D printing or Additive Manufacturing (AM) was originally born as a mono-material technology. And, nowadays, most of the applications are still using only one material.

A brief classification of the AM technologies, according to this subject, is given next:

- "Native" mono-material AM technologies: VAT Photopolimerisation, Powder Bed Fusion, Binder Jetting and Sheet Lamination have homogeneous base material. This base material is spread over an exposed flat surface, therefore being consolidated layer-by-layer.
- "Feasible" multi-material AM technologies: Material Jetting, Material Extrusion and Direct Energy Deposition have more room to move towards multi-materiality, as long as different materials can be added to every single layer forming the parts. But, in fact, most of the present applications are limited to mono-material parts.

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The first group of the aforementioned AM technologies are generally those which were first invented in late 80' and first 90' of the XXth century. Attempts to reach multi-material parts using them has been made, but have been limited to the research field. Anyway, very interesting outcomes have been made, and some of the most relevant results that set the state-of-the-art for every AM methodology are summarized here for three of these technologies:

- VAT Photopolimerisation: a layer of homogeneous photopolymeric resin is solidified. Choi et al. [1] detailed that multi-material parts can be obtained by combining different resins using a rotating vat carousel system. Different vat is needed for each material, and time and cost is huge compared to others. Multi-material parts can be obtained combining VAT photopolimerisation with material jetting drop-on-demand technology using conductive inks [2].
- Powder Bed Fusion (PBF): this technology has been associated with a homogeneous powder, commonly PA11 and 12 for polymers, and also for a limited range of metals like titanium alloys. So, multi-materiality has been obtained by mixing fusion-compatible different materials. A wide range of possibilities has been explored by mixing polymers with ceramics [3], metallic composites as Fe and graphite [4] or Fe, Ni and TiC [5] and ceramic composites as Si-SiC [6]. Research about the characterisation of multi-material powder can be found in Roure et al [7].
- Binder Jetting: is based on adhesive deposition in drops selectively over a layer of powder (plaster, sand, concrete, metal...), and controls the composition of the product based on projecting different materials (inks) using different nozzles. Then, the amount deposited can be controlled so it varies progressively and gradually obtains materials with variable properties [8].

The second group of AM technologies, more capable to offer multi-material parts, are being also mostly used nowadays for mono-material 3D printed parts, as long as they are commonly replicating mono-material products. And that is the main point: as long as traditional industrial production systems are mono-material oriented, there is a sort of "cultural resistance" to image how products can be re-though taking advantage of the freedom of design inherited to AM, not only about freeform geometry but also about the multi-material possibility. That is why the present manuscript explains the need for multi-material 3D printing and focuses more thoroughly on Material Extrusion technologies.

2. Need for Multi-Material 3D Printing and Technologies

AM has a lot of potential but has not yet been fully explored, and access to the creation of multi-material products is an example of it. In the real world, most of the objects around us are multi-material, and it is thanks to this feature that they can perform their function appropriately in different industrial sectors as far away as automotive, food or health [9-11].

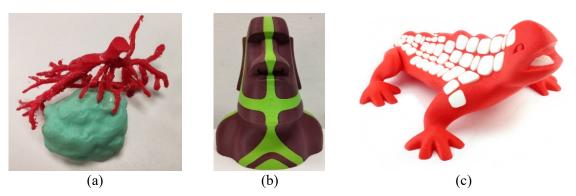


Figure 1. (a) Tumour with blood vessels using TPU (Thermoplastic polyurethane) filament (b) Demonstrator using PLA (PolyLactic Acid) (c) Salamander using PLA.

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The most obvious progress has been made in the field by incorporating colour into the 3D printed parts (see figure 1), but one of the most interesting areas is the introduction of materials with different rigidities, stiffer and softer areas, with differentiated values of mechanical strength and viscoelasticity.

The latter has been mainly achieved thanks to the material jetting technologies, since it can 3D print samples with different mechanical and viscoelastic properties thanks to the concept of voxel 3D printing. However, its price is a limitation, being mostly used by large companies.

For Multi-Material Jetting in the more accessible desktop 3d-printing sector, several developments combining it with other technologies have been made, such as digital light processing. For example, CIM UPC developed another hybrid 3D printer by the combination of DLP with Material Jetting under the NHIBRID project. By this way, multi-material parts can be manufactured for electronic devices.

For Multi-Material Extrusion, the most common way to perform multi-material parts is the direct use of different liquids (DIW technology) and/or filaments (FFF technology) that, under multiple strategies, ends up obtaining an object [12], as is presented in the next point. On the other hand, CIM UPC has the know-how for manufacturing this type of machines in different projects. For instance, in the medical field, a hybrid multi-material 3D printer was developed for the manufacture of surgical planning prototypes. This machine combined both DIW and FFF technologies (see figure 2).

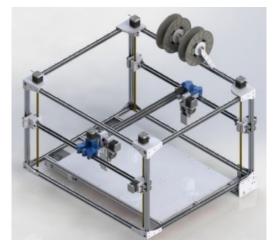


Figure 2. CAD file of a multi-material 3D printer able to assembly a cavity-pump print head in one of the axes.

3. Material Extrusion: Research on Desktop 3D Printing New concepts

The concepts represent, in many cases, scientific problems not addressed to date, and their initial embodiment in the form of prototypes is a way of verifying the difficulties that would be encountered in the transition from research to reality.

These new concepts explored were introduced in a previous communication by some of the authors [13]. In that past communication, the focus was placed on trying to mimic living tissue by means of multi-material 3D printing. Here, the research is not narrowed to a possible market application, but opened to the general purpose of multi-material 3D printing.

In brief, these are the concepts: (1) revolver print-head, (2) filament convergent nozzles, (3) filament mixing print-head, (4) polyurethane print-head, and (5) 3D printing with UV silicone.

3.1. Revolver print-head

A revolver print-head design has been made that places only one nozzle at a time in the printing plane, so that the nozzles that do not act are not rubbing against the layer of material already made. This design is similar to the one allowing the change of objectives in a microscope or the revolver turrets of a CNC

lathe (see figure 3). One of the advantages of the solution derived from the research carried out is that it uses a single extruder mechanism even though there are several filaments.

The experimental concept developed of revolver print-head has been able to make impressions, completing tests with parts of one and two colours with satisfactory results. With a correct adjustment of the printing parameters and the implementation of some improvements, it is concluded that this is a way to have multi-material prototypes within the range of materials and colours that the FFF technology allows

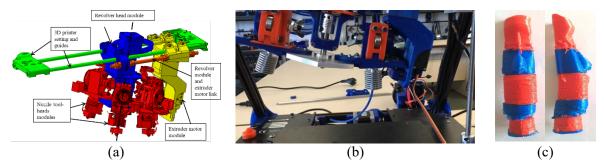


Figure 3. (a) Revolver print-head design concept (b) demonstrator [12] (c) firsts 3D-printings using two materials

3.2. Filament convergent nozzles

The key point in this concept is the ability of the print-head to convey different materials without mixing them, therefore having the capacity to deposit them in different proportions. The print-head does not need any mixing chamber, making any purge action unnecessary when changing material. Independent nozzles place the extruded material at the same point, simulating by juxtaposition the mixing effect (see figure 4).

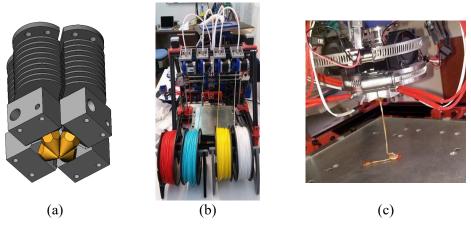


Figure 4. (a) CAD design of the filament convergent nozzles concept **(b)** Overview of the filament feeding system and 3D-printer with filament convergent nozzles concept **(c)** All the nozzles extruding simultaneously different materials [12].

The option of not mixing the filaments inside the print-head before its deposition is considered of high interest in order to make combinations of materials (PLA, ABS, Filaflex®, etc.) so that they are virtually mixed. Each nozzle is a true print-head in fact, so extrusion parameters (like temperature) are controlled separately, attending the needs of every material and achieving different physical properties within the final part. Making a comparison with metals, it would be like getting together non-alloyable materials as it is made in the sintering manufacturing technology.Performed tests indicate that technological viability of this concept may be more difficult to reach than the previous revolver print-

head concept, as long as there is work to be addressed about the reliability of the nozzles tilting to make them converge at one point due to filament jamming in the final section, due to the change of exit angle. Also, it has been stated that the quality of the print can be compromised depending on the direction of advance of the head due to their non-vertical extruding axis (see figure 5).

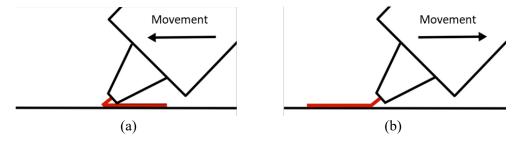


Figure 5. One of the results of the filament convergent nozzles concept was related to the quality of the print. (a) Deposition of filament in the direction of the nozzle, combing the deposited layer and compromising its quality. (b) Deposition in the opposite direction, which does not affect the deposited layer [12].

3.3. Filament mixing print-head

Compared to the two previous concepts, this concept simplifies the requirements of the printing process, making the print-head simpler: filaments are "poured" into a single print-head, being extruded through the same output nozzle (see figure 6a). Therefore, multimaterial parts can be made by the contribution of "pure" or "mixed" filament.

Another important question when going through this concept is about how material mixing is performed inside the print-head. Two possibilities, passive or active mixing, arose and were handled. So, two different print-head prototypes were designed and materialised.

Passive mixing revealed poor quality extruded material. In one of the tests performed, two materials were extruded simultaneously, in some cases using the same speed and in others assigning different speeds for each of them. Although the quality of the mixture has mostly been considered acceptable, in some sections strip distinctions of the two materials in the direction of extrusion were signalled. Without an active mixing system, in some sections the mixture was not properly mixed and one half of the filament of each of the extruded colours can be clearly distinguished, as it can be appreciated in figure 6b.

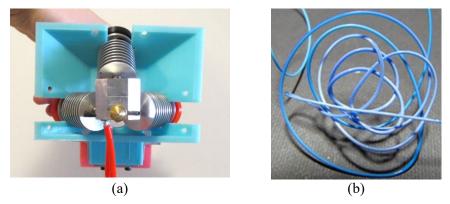


Figure 6. (a) Filament mixing print-head for 3 materials (passive mixing). **(b)** Extruded sample of two materials (blue and white PLA filaments) [12].

So, active mixing systems have been explored too. An embodiment for this concept integrates a mixing shaft, which will be driven at a low, continuous and regular speed, in order to obtain the desired homogeneity in the mixture. The upper part, conveniently sealed to prevent filament reflux, is designed

to ease its connection with a stepper motor by means of a flexible coupling, while the lower part, determines how the mixture will be carried out: its prismatic shape leaves spaces (four sub-chambers) between the shaft and vertical duct of the head where the filaments converge.

To sum up about this wide research performed over this concept, these statements can be made:

- The experimentation of active and passive mixing solutions in the print-head has benefited from being one of the most studied solutions within the open source developments of FFF technology. However, maybe this concept is not capable of performing 3D printing multi-materiality to the desired level.
- The need to program an effective control of the mixtures has forced to advance the research using calculation and simulation tools to find the parameters of extrusion that were suitable for correct 3D printing. CAE simulations indicated that, for a good mixture of the different filaments, it will be of interest to work with a rotation speed of the mixing motor between 5 and 15 min⁻¹.
- Difficulties encountered in experimentation have been more related to improvements needed in mechanical design than to issues inherent to the concept.

3.4. Polyurethane print-head

Experimentation over this concept has been focused on polyurethanes obtained by reaction of two components (polyol and isocyanate), thus making the reaction impossible because of the time of mixing and 3D printing. It is a well-known material for the research team, given that at CIM UPC, where the tests have been performed, polyurethane is used as a raw material for prototype pre-series, casted in vacuum silicone moulds made by means of a 3D printed master model. It is possible to present them in different colours, adding pigments to the base materials, densities (possible injection of air), hardness (also adding additives), making polyurethane theoretically capable of forming a functional "multi-material" 3D-printed part.

In this concept, tests of manual deposition by superposition of two layers were performed to see if setting times are short enough for a layered manufacturing process, that is, waiting time to 3D-print every layer. Temperature was also an important factor to monitor, in order to determine the need for a controlled-temperature closed volume on the 3D-printer between 50 and 70°C. After the tests, it was seen that polyurethane was an unattractive material for 3D printing due to its very long setting time (see figure 7).

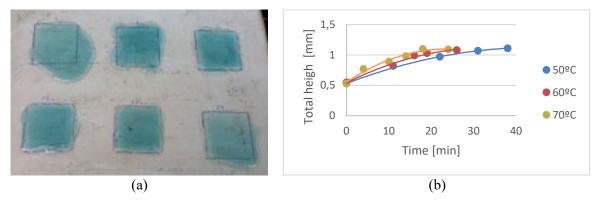


Figure 7. (a) Deposition tests of two layers of polyurethane at 70°C, using different deposition times for the second layer. **(b)** Layer height with respect to time and different temperatures [12].

3.5. 3D printing with UV silicone

The first step related to this concept was to search the market availability of UV-curable silicones, and to know their basic characteristics such as the necessary time of exposure to UV-light and the range of hardness of the product once catalysed, in order to accept silicone as "multi-material" if this range is

open enough. So, the exploration with curable silicone with UV-light as a material for 3D printing began, and simple tests of drop deposition and exposure to UV light were carried out which were satisfactory.

Starting from a BCN3D+ open source desktop 3D-printer with the Paste Extruder kit, the challenge was to modify it to incorporate a support in the print-head to fix a laser pointer. Once the equipment was ready, experimentation began. The final tests with UV-curable silicone gave good results, almost eliminating the free exit by gravity in the dispensing, possibly due to the effect of the higher viscosity of the silicone with respect to previous trials with epoxy resin used to calibrate the system (see figure 8). The instantaneous solidification of the deposited drops made it possible to avoid the "spill" effect, but in the tests it was necessary to speed up the process to avoid blocking the syringe.

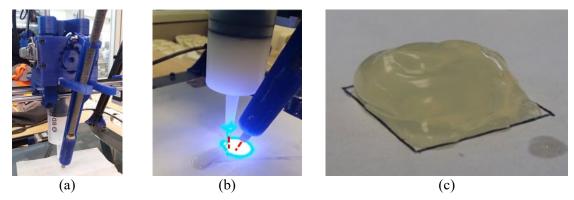


Figure 8. (a) 3D-printer adapted to 3D print with silicone. (b) UV laser at work, catalysing silicone. (c) First attempts to construct a 3D-printed cube with UV-curable silicone [12].

4. Conclusions

The present work illustrates not only the under-developed path to 3D printed multi-material parts using material extrusion technologies, which are cost-effective [14,15], but also the tiny level of application amongst the different AM technologies, and a significant research work developed in the 3D printing desktop sector.

To conclude, a more concise assessment of the multi-material 3D printing desktop concepts explored:

- Revolver print-head: suitable solution if the multi-materiality is limited to filaments, using up to five different materials. It is considered impossible to combine FFF (filaments) and DIW (liquids) print-heads in a single rotating structure.
- Convergent nozzles: suitable solution if the multi-materiality is limited to filaments, using up to four materials. It would also make sense to have a different print-head, still under exploration, for UV-catalysed silicones of different hardness and/or colour, therefore making possible to obtain parts with a wider range of different values of physical characteristics.
- Filament mixing print-head: it is the most common path found in the state of the art. Compared to the first two concepts, it is less difficult to materialise. To achieve a wide multi-material solution, differentiated mixing print-heads can be developed using different ranges of materials.
- Polyurethane print-head: there are still relevant issues to overcome, related to the material (difficult cleaning, concerns about VOC and labour safety, long catalysis time) and for the fact that there are other "configurable" materials, as for example silicones, that can better handle these problems and offer more range of values (e.g. reach lower hardness values).
- 3D printing with UV silicone: catalysis time is improved, making possible an acceptable buildability on a 3D-printing. Anyway, similarly to the previous concept, there is still a lot of research to do, especially in the chemical field related to the development of new UV silicones in order to generate different physical properties that allow multi-material parts made by different graded silicones.

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