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Short Communication

3D Printing Filaments with Electrical Properties: From the Fundamentals of Additive Manufacturing to Recent Trends

Evando S Araújo[1]*, Georgenes MG Silva[1,2], Anilton CS Melo[1], Evandro A Silva[1], Francisco F Silveira[1], Mychel MF Santos[1], Sofia R Mendes[3], and Pedro M Faia[3]

[1]Research Group on Electrospinning and Nanotechnology Applications, Department of Materials Science, Federal University of São Francisco Valley, Brazil

[2]Federal Institute of Education, Science and Technology of the Sertão Pernambucano, Brazil

[3]CEMMPRE - Electrical and Computer Engineering Department, FCTUC, University of Coimbra, Portugal

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***Corresponding author:** Evando S Araújo, Research Group on Electrospinning and Nanotechnology Applications, Department of Materials Science, Federal University of São Francisco Valley, 48902-300, Juazeiro, Brazil

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Abstract

Additive Manufacturing (AM) can be established as a differential production technique in terms of reliability, economy, repeatability, and scalability for industrial purposes. In this context, the Fused Filament Fabrication (FFF) based printing technique has been providing additive manufacturing applications in several areas of knowledge, based on the production of functional composite filaments, including those of the electrical conducting type. In this sense, this communication presents fundamental aspects of AM, with a focus on FFF 3D printing, and recent applications of conductive filaments in Materials Science area.

Introduction

The industrial production mode of durable and non-durable materials, such as parts, tools and machines, with well-defined three-dimensional geometries for various applications, is directly related to manufacturing engineering techniques. Turning, casting, welding, molding and forming of parts are some of these so-called conventional and widely explored forms of production for the manufacture of objects from metal, ceramic and/or plastic-based raw materials [1,2]. Many of these techniques use the removal of excess volume from the given material so that the designed three-dimensional model can be created [3,4]. In recent years, new manufacturing models have been explored, such as the use of the additive manufacturing method, which considerably reduces the loss of raw materials during the manufacture of objects.

Additive manufacturing (AM, popularly known as 3D printing) represents a set of technologies that allows the creation of three-dimensional objects using digitally designed models [5]. These objects are printed on equipment's called 3D printers, from successive depositions of layers of a certain molten material of interest (for example, polymer, metal, ceramic, composite) on the printer's working surface, until the volumetric object is fully built [6].

This method of manufacturing three-dimensional objects is commonly used in the production of prototypes and models in the areas of architecture and engineering (models for structural analysis, automotive and aeronautical parts with improved properties, flexible and personalized electrical circuits from emerging materials, etc.), in biomedicine and bioengineering (personalized anatomy, prosthetics, tissue engineering, bone regeneration, drug delivery systems, among others), and education (materials dispositive to improve the teaching-learning process). In fact, 3D printing has been gaining enormous interest both in academia and industry due to its potential use for improving the properties of built parts, production quality with repeatability and precision, in addition to reducing production costs in several areas.

AM production has as differentiating factor in relation to other production techniques, the possibility of manufacturing personalized objects with higher process execution speeds, even with complex geometries, which would be difficult to obtain using other manufacturing techniques. In the literature, several 3D printing techniques are found, the most common being Stereolithography (SLA), Direct Light Processing (DLP), Selective Laser Sintering (SLS), Direct Metal Laser Sintering (MDLS), bioprinting, Selective Laser Melting (SLM), Fused Filament Fabrication (FFF) or Fused Deposition Modeling (FDM) [7]. The FFF 3D printing technique stands out among the printing techniques mentioned because it is the most conventional, low-cost, and has applications in several scientific fields. Therefore, this last technique will be presented below, focusing on the development of filaments with electrical conduction properties for emerging applications in Materials.

Basic of FFF 3D printing

FFF 3D printing technique makes use of cylindrical millimeter filaments of thermoplastic polymers, which are melted at a suitable temperature (by the heating system of a compatible 3D printer) to form three-dimensional parts. The molten polymer filament reaches the printer's millimeter extruder nozzle and then, is deposited on the equipment's working surface. The part construction process continues with the overlapping of layers of the material until the designed three-dimensional object is complete, with the polymer returning to the initial solid phase.

Generally, the technique is used to produce conventional objects with specific mechanical resistance characteristics, depending on the choice of polymer. More recently, the potential of the technique has been on its to produce functional objects, from polymer composites with additives and/or functional particles. The technique is also widespread in areas of basic and applied science due to the versatility of polymeric materials and their composites used, and to the possibility of producing processed materials with different desirable physical properties (mechanical, thermal and electrical) [8].

Prior to the printing process, it is necessary to arrange a polymeric filament with the properties required for the application. These polymeric filaments are produced using hot extrusion technique. In this process, grains of the polymer of interest (available with or without additives, and/or functional particles) are inserted into an extrusion machine to be melted at a suitable temperature and are then extruded in the form of cylinders with diameters of 1.75mm. or 3mm, compatible with available printer systems. The most common polymeric materials for the production of polymeric filaments are Polyamide (PA), Polyethylene Terephthalate Glycol (PETG), Acrylonitrile Butadiene Styrene (ABS), polyethylene terephthalate glycol and Polylactic Acid (PLA). More recently, unconventional filaments have been developed resulting from the mixture of these polymers and/or the incorporation of functional particles into the filament volume for specific applications, such as the production of personalized drug release devices, functional prostheses and the production of flexible electrical circuits.

Properties such as malleability, biodegradability, environmentally friendly material, greater flexibility and lower melting point have been preferable for the use of thermoplastic polymers in FFF 3D printing for emerging applications. The production of 3D printing filaments with electrical conduction properties, as well as their potential applications, fall within this context and will be discussed below [8].

FFF 3D printing filaments with electrical conductivity

Some types of unconventional filaments are known to have electrical conduction properties resulting from the introduction of micrometric and submicrometric conductive particles into the volume of the polymer matrix. These special filaments can be used to produce flexible and/or customized electrical circuits/devices with direct application in the area of electronics. In the literature, it is possible to find these composite filaments from the addition of gold, silver, copper, aluminum, and titanium nanoparticles, and carbon-based materials such as carbon nanotubes, graphite oxide, graphene oxide and reduced graphene oxide. These conductive particles can be obtained from chemical synthesis processes highlighted in the literature. In addition to electrical conduction, personalization and/or miniaturization of devices, the use of these composite filaments with conductive particles becomes attractive to produce electronic materials due to the lightness and flexibility of the final composite produced from the constituent materials. Some of the most recent advances in this area of research are summarized below.

Zhang et al. [9] produced flexible 2D and 3D electrical circuits by FFF 3D printing, from PLA/reduced graphene oxide (rGO) composite filaments. To prepare the filaments, the authors initially mixed PLA and rGO at 160 °C in a melt mixer, without adding solvents. Then, the material resulting from the mixture was cooled, cut into pieces, and taken to a mini extruder so that PLA/rGO filaments with a characteristic diameter of 1.75mm could be manufactured. The conductivity obtained for the PLA/rGO filaments was approximately 5S/m, using 6wt% of rGO. The authors concluded that this composite filament is a potential candidate to replace copper, as the circuits printed with it have compatible conductivity for various applications of electrical materials.

Dul et al. [10] produced ABS filaments containing conductive Carbon Nanotubes (CNT) with piezoresistive properties for application in deformation monitoring. The authors highlighted the ease of the filament manufacturing method by extrusion at laboratory scale. The parts produced from filaments with 6wt% CNT were mechanically tested, and the piezoelectric properties were analyzed during short- and long-term periods. Variations in electrical resistance of the ABS/CNT composite devices were observed in several cycles and associated with the percolation network of conductive particles. The stability of the resistivity results in the temperature ranges from −25 to +60 °C was used as a signature of the mechanical stresses performed.

More recently, Dembek's group [11] showed the importance of adjusting printing parameters to minimize the resistivity of 3D printed electrical conductive structures, produced from commercial PLA filaments containing carbon nanotubes as conductive particles. The authors concluded that the printing process with higher temperatures, larger printing nozzle size, and optimum layer height, increase conductivity of the final three-dimensional printed parts. For example, the lowest resistivity in the printed samples was approximately 3Ω.cm, with an extrusion temperature of 230 °C, a 1mm nozzle, and a 0.7mm layer height. Future studies with other parameters such as printing speed and part filling density, and using other materials, are also suggested by the authors.

Verma et al. [12] contributed to this field of research by studying the Alternate Current (AC) conductivity of filaments resulting from the dispersion of Multi-Walled Carbon Nanotubes (MWCN) in a Prolipropylene Matrix (PPR). The authors observed that the good dispersion of MWCN in the filament bulk resulted in better mechanical and electrical properties of the printed parts, compared to the use produced with the polymer without the addition of conductive particles. The AC conductivity tests were performed in the frequency range of 20 to 107Hz. The results showed a significant increase in the conductivity of the parts throughout the frequency excursion, with a variation from 10-13 to 10-7S/cm for the samples with 2wt% MWCNT. In addition, the samples with 2wt% and 4wt% MWCNT showed frequency-independent conductivity up to 104Hz, but the conductivity showed a frequency-dependence increase above this value. The authors highlight that conductivity variations in disordered materials such as those studied, are generally explained by the polarization effects between clusters of particles and the anomalous diffusion of charges along these conductive sites. In fact, the results show a percolation of the electrical conduction in the composite materials tested, which explains their conductive behavior with frequency. Above the percolation threshold, at low frequencies, conductivity is governed by the effect of random conductive paths that are formed between MWCNT particles in the bulk of the insulating matrix. At high frequency, the increase in the capacitive effect of the clusters becomes evident and the conductivity of the samples increases.

The work of Ghosh and collaborators [13] studied the (photo) electrocatalytic activity of a series of molybdenum disulfide (MoS_2)/C/PLA composite filaments, where C represents conductive fillers of carbon materials such as graphite, activated charcoal, and MWCN. The devices produced from the conductive filament showed excellent photocatalytic activity in the red wavelength region, and high areal capacitance and cyclic stability for energy storage applications. For information, the best results obtained with the printed devices involve capacitance of up to 381mF/cm^2, current density above 1.8mA/cm^2, and 92% efficiency in cyclic voltammetry studies after 6000 cycles.

Another recent study showed the improvement of electrical conductivity of PLA matrix filaments with MWCNT fillers influenced by the addition of Lignin as a bio dispersant [14]. Electrical percolation of the filament with lignin at 1wt% was obtained with 5wt% of conductive material, with a variation from 10^{-7} (below threshold) to 10^{-1}S/cm, without significant changes in its mechanical properties in comparison with the observed for filaments without dispersant. In addition, the authors report that the use of lignin is more advantageous than the use of Polyethylene Oxide (PEO) dispersant in relation to the material processability during the printing process, and to the adhesion between layers of the printed object.

It is possible to mention other works on conductive filaments that focus their attention on the development of new personalized biosensors. Just as an example, we cite the example of a filament easily produced from a mixture of PLA and graphite for the development of the basis of an electrochemical biosensor for detecting SARS-CoV-2 virus [15]. The PLA/graphite filaments were used to manufacture the electrodes of the electrochemical cell, which underwent a chemical process so that the virus antibodies could bound to the surface of the device and thus provide the detection of the virus's biological agents. The electrical conduction properties of the composite filament, combined with the surface chemical modification carried out, were essential to obtain a personalized device, with repeatability of the linear detection signature of the tested virus load.

Conclusion

The Additive Manufacturing (AM) production technique was presented and discussed in relation to conventional production methods, focusing on FFF 3D printing and manufacturing polymer composite filaments with electrical conduction properties. The development of these unconventional composite filaments containing conductive particles has been widespread both in academia and in industry today, as a viable and innovative alternative to produce electrical devices for on demand applications. From the search on recent works about the topic, it was possible to show the importance of studies on filaments with these characteristics for the development of new functional products and processes in the electronic materials area.

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Conflict of Interest

The authors declare no conflict of interest.

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