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Trends of using polymer composite materials in additive manufacturing

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Abstract. Additive manufacturing (AM) technology is nowadays one of the advanced manufacturing technologies used to convert three dimensional product data to physical objects without any special tooling. The emergence of the technology has made production of complex geometries having consumer demands for more customized products and services. Though the underlying technology was initially intended for rapid prototyping, the technology has recently getting wider attention in diverse industrial sectors using different material types commonly polymers, metals, ceramics, and other compatible researched materials as input. Because the technology processes the input materials with no or very low wastes, the use of polymer composite materials leads to a no waste manufacturing system. This article aims to review and analyse the opportunity and existing challenges for AM technology in processing polymer composites and provide an insight on the state-of -the art applications. The paper provides an overview of the materials and processes currently available for this novel production technology to fill the industrial needs for engineering application. In addition, closely reviewing published research works in the area, the existing research gaps are identified for possible future research in the field.

1. Introduction

Additive manufacturing (AM) is one of the emerging manufacturing technologies that is used to form three dimensional products by adding materials consequently layer by layer at a time through the bottom-up approach. Due to the way parts are produced, it is often referred to as 3D printing. Thus, in this article, the words additive manufacturing and 3D printing are used to imply the same technology. The concept Rapid Prototyping (RP) is also, sometimes, synonymously used though Rapid prototyping is the earlier development of the technology that allows engineers to understand and explain their design through computer-aided design to conceive as a technique for production of physical models or prototyped parts from virtual model that enhance the efficiency and reliability of production system. The improvement for 3D part printing process and advancement of material processing attracts market to prefer this technology that helps to lead the transition of old production system towards additive manufacturing system to be used as user interfaced parts production method are among 21st century technology to fulfill basic human engineering needs which is under alarming growth [1].

Emerging as a rapid prototyping tool, AM is timely mechanism for adoption flexible manufacturing system (FMS) that can be applied to diverse material types including thermoplastics, metals, concretes and ceramics. However, 3D printing predominantly targets the production of polymeric parts and models it prefers suitability associated with photo polymerization and availability of polymers. This is



partly due to the fact that polymer composites are easy to reprocess the wastes and convert into new raw materials for AM.

The 3D printing technology covers a wide range of technologies and gives a clear advantage over the existing manufacturing systems, especially for complex geometric parts, precision resources, short manufacturing lead time and customized products. According to Attaran [2], only in the year 2014, the key players in the manufacturing industries spent nearly about \$3 billion in additive manufactured components. These include industrial machines 18.5%, consumer products 18.0%, automotive industry 17.3%, medical/dental sector 13.7%, aerospace industry 12.3%, and others 19.1% such as academic institutions, the military sector, architecture, etc.. The global sales of AM machines, related materials and services have also shown an average annual growth of about 33% in the period 2013 – 2015 [3]. The same study indicates that AM technology is largely used to produce functional parts and prototypes for fits and assembly. In terms of the printing materials, polymer-based materials account for the largest share, particularly in the production of medical implants [4].

Though the technology emerged in the product design and manufacturing environment not more than three decades ago, it has already left its footprints with a potentially huge impact to transfer the manufacturing method to a real automated, flexible, fully customizable and digitally controlled process. However, there still exist a number of bottlenecks including reluctance of the manufacturing sector to adopt as a viable manufacturing method. In addition, several process specific characteristics, machine related constraints, materials and achievable qualities both as dimensional and geometric deviations and surface qualities influence the opportunities that this technology can provide [5].

The aim of this paper is to conduct a review of the opportunities and challenges associated with additive manufactured functional parts using polymer composite materials. Following this introduction section, the paper consists of 5 sections. The next section presents the key polymer materials including ceramics and cement based composites used in AM based part fabrication process. In addition to some of their mechanical properties, their application areas, benefits and main limitations are highlighted. Then the trends in industrial application of AM as functional part fabrication method is presented, and followed by the outlook for the technology roadmap and standardization works for AM application. Based on the conducted review and analysis, some observed research gaps are highlighted and then drawn conclusions of the study are summarized.

2. Polymer composite materials for AM

2.1. Mechanical and functional properties of additive fabricated polymers

As mentioned earlier in this article, polymers are not only the first material types used for rapid prototyping using the layer by layer fabrication approach; they are also preferred as commonly used materials for the AM industry. This is primarily because of the diversity of polymer-based composites and the simplicity to adopt them to different AM processes. Polymer-based materials for AM are available as thermoplastic filaments, reactive monomers, resin or powder. They are marketed as ABS, polyamide (PA), polycarbonate (PC) and polylactic acid (PLA), and thermosetting powders such as polystyrene, polyamides and photopolymer resins, are the most common types of polymers for additive fabrication [6] by many AM machines such as stereolithography apparatus (SLA), selective laser sintering (SLS), fused deposition modelling (FDM) and 3DP inkjet printers.

Some of the benefits of fabricating polymer composites using AM technology include the ability to customize complex geometries with high accuracy [7]. Contrary to the attractiveness of polymer composites with respect to their diversity and ease of adoption, their inferior mechanical properties have impact on their application for load carrying structures. As a result, a large size of research and development has focused on developing various methods and materials for prototyping purposes. In order to overcome the limitations of lack of mechanical properties of polymer composites, which are limited to prototyping purposes, the reinforcement of polymers with fibres and nano-materials has been introduced in recent years. These efforts are aimed to enhance the mechanical properties of the printed products to be used as load-bearing or functional components [6, 8]. In this regard, nylon based polymer composites i.e. PA is one of the widely used polymers for SLS operations because of its easy melting using lasers, compared with other forms of polymers [9 - 11]. Other polymers such as

polystyrene, thermoplastic and elastomers are also well known as suitable materials for SLS process [12]. Among the polymer composite materials processed by FDM technology, ABS is most popular [13] for general applications, while a recent developed high performance polymer called ULTEM has attracted interests [14, 15] for additive fabrication of parts that require higher strength, durability, toughness, high tensile and flexural strength including resistance to heat and chemicals.

Due to its popularity as industrial class additive fabrication for polymer composites, FDM is recently under extensive investigation. Due to the inherent poor mechanical properties of the polymers, however, FDM process itself suffers critical limitations such as unpredictable dimensional accuracy and surface quality. As several fabrication parameters, such as raster angle, contour width, air gap, etc. (Figure 1 [15]), influence the accuracy and surface roughness, there exists no reliable mechanism to control the accuracy. For instance, a recent study conducted in our 3DP laboratory on 3D printed parts using FORTUS 450mc, with 45° and 90° orientation, show significant variation of the surface quality in terms of all measured parameters.

As can be observed from Table 1 [16], the surface roughness has better qualities in the 90° orientation while the 45° orientation provides better flatness accuracy. Though geometric complexity, lack of mechanical strength and functionality are often mentioned to be the forefront challenges limiting the wide applications of additive manufactured polymer products, the polymers in liquid state are the most widely used materials in AM industry due to their low melting point, low weight, low cost and processing flexibility. Combining various materials for achieving desired mechanical and functional properties is a promising way to solve the challenges in AM of polymer products. Therefore, development of composite materials that are compatible with the available additive fabrication technology has recently attracted tremendous attention. Short fiber-reinforced polymers, in particular, are most realistic for FDM based process and result in moderate improvement of mechanical properties [17, 18].

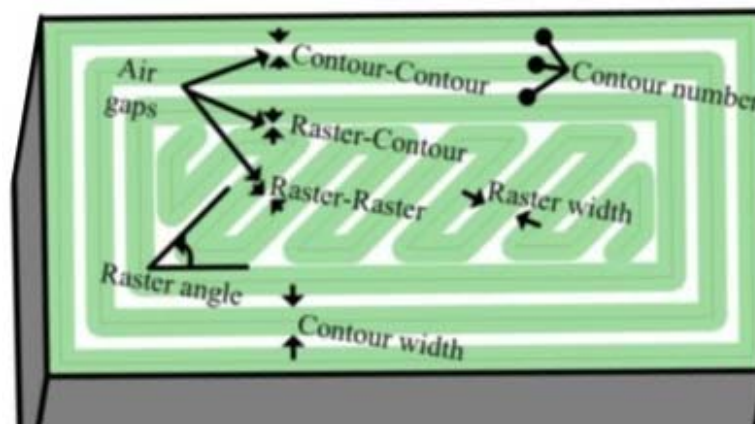


Figure 1. Process parameters for FDM technology.

Table 1. Values of surface quality measured in two different print orientations.

Print orientation		45° [μm]	90° [μm]
Surface finish parameters	S_a	15.389	14.113
	S_q	20.339	18.290
	S_v	76.964	86.888
	S_z	135.438	140.677
	S_{sk}	-0.370	-0.227
	S_{ku}	3.312	3.249
	FLT_t	135.438	140.677

Notations: S_a = Average height; S_q = Root-Mean-Square height; S_v = Max. valley depth; S_z = Max. height; S_{sk} = Skewness; S_{ku} = Kurtosis; FLT_t = Flatness using least squares reference of the selected area.

The progressive developments in AM technology has enabled processing of a wide range of materials from chocolate to advanced multifunctional materials. Materials can be processed in the forms of filaments, wire, powder, paste, sheets and inks. Among these, polymer composites are considered as the most common materials applied in aerospace, automotive, sports, medical, architectural and toy industries. While those used in FDM technology are in the form of filaments, those in powder form or auxiliary binders in the powder-bed processes and resins in liquid state are used in SLA machines [6].

2.2. *Ceramics for additive fabrication*

Additive manufactured ceramics have created a trend to tailor-design materials with a high strength to weight ratio and facilitated the creation of complex ceramic lattices for many applications [6]. In particular, strong and versatile ceramic scaffolds with complex shapes for tissue engineering is one of the main applications. The possibility to embed biosensors in medical devices or human organ has significantly advanced biomedical sector [19]. Ceramic materials for additive fabrication are in the form of powders that are sintered using laser or bond together via an auxiliary adhesive or in the form of ink for ink-jet printing through suspension of ceramic particles at high sintering temperature followed by post-treatment. The main challenge is the limited availability of ceramic materials for additive fabrication using currently existing methods, relatively poor dimensional accuracy and quality. On the other hand, additive fabrication of ceramics provides better control over the microstructure and composition of the part. Therefore, research and development in the optimisation of AM technologies, as well as the expansion of materials selection for fabrication of ceramics are opportunities that are yet to be exploited [20].

The main application area, pros and cons of ceramic and polymer materials for AM is presented in Table 2. Further details are also available in the literature [21].

Table 2. Ceramic and polymer materials, Pros and cons for AM.

Material	Main applications	Benefits	Limitations
Polymer composites	Aerospace, sports, medical, toys and architecture	Allow fast prototyping, cost effective, fit for complex struct., cost effective and customized product.	Mechanically weak, limited availability incl. reinforcements, anisotropic
Ceramics	Biomedical, aerospace, automotive & chemical industries	Control of lattice porosity and printing complex structures and scaffolds for human organs, reduced fabrication time, etc.	Limited avail. of printable ceramics, inaccuracy and poor surface finish, and inaccuracy if not post processed.

2.3. *Concrete based composites*

Concrete is the most ancient and widely used man-made material (composite) particularly within construction and infrastructure sector. Employing the concept of the deposition process from other small size processes like FDM and 3DP technologies, the first cement-based additive fabrication [22] attempted to glue sand layers using Portland cement paste. The process can be considered as an expansion of 3D printing to large-scale application [23] in the domains such as design, construction and architecture, using cementitious materials [24 - 26].

Though the adoption within this industrial sector has been at poor progress, its benefits such as mass-customisation, building process automation without the need for formwork and the like has created interest in the field. The main techniques used to additively fabricate concrete is the extrusion process, though the powder-bed method has been also researched well. Many factors influence the suitability of the concrete type both in terms of its simplicity to extrude, shape stability and dimensional accuracy after fabrication. The formwork-free fabrication may not fit for self-compacting concrete as it may not hold its shape. Furthermore, other problems such as anisotropic mechanical properties and poor inter-layer adhesion are the main challenges that need to be addressed. Despite

these challenges, freedom of design and opportunity to build complex and lightweight structures are promising [1, 6].

2.4. Multimaterial printing and process control

One of the key motivation for multimaterial printing is in order to get multifunctional properties. For instance, current AM technologies can provide a multifunctionality of additive manufactured parts by composite materials while providing their required composition property [6]. For such composites, though there are some existing limitations, high-pressure jetting systems can combine only polymers with good flow ability and similar curing temperatures and extrusion-based methods. While FDM process can couple only materials with similar melting temperatures, other AM technologies, such as direct energy deposition (DED), allow for the combination of metal alloys or ceramics, and as a result, more materials can be extruded at the same time. However, additional care must be taken to avoid the formation of unwanted phases between two materials in contact [6].

Multimaterial AM has been successfully achieved in some powder-based technologies using powders with different melting temperatures. In such cases, issues related with remelting or degradation of materials with lower melting points can be encountered adjacent to the powders with higher melting points. Though performance prediction is expected to be complex, the development for optimization through numerical modelling approaches can assist with the multimaterial manufacturing of 3D parts within their designed multifunctional purpose.

3. Development trends of AM as industrial process

The adoption of AM and other advanced manufacturing technologies is heralding the future manufacturing process in which value chains are shorter, smaller, more localized and customized, more collaborative, and provide sustainability benefits. Compared with traditional subtractive processes, the following potential benefits stand out [4]:

- Improved resource efficiency.
- Extended product life.
- Reconfigured value chains.
- Wastage minimization of costly metals like titanium.
- Advancing design for assembly method to improve properties and minimize cost.
- Highly customized products to individual needs.

There are sufficient indications that make us believe that AM continues to be implemented into the modern industrial processes as well as human livelihood as this technology continues to develop, broaden its scope of applications and continue to be available at reasonable costs. Only within the last three decades, the AM related industry including the technological and material developments and related services have shown an exponential growth. As shown in Figure 2, almost a double growth is observed only within two years [3].

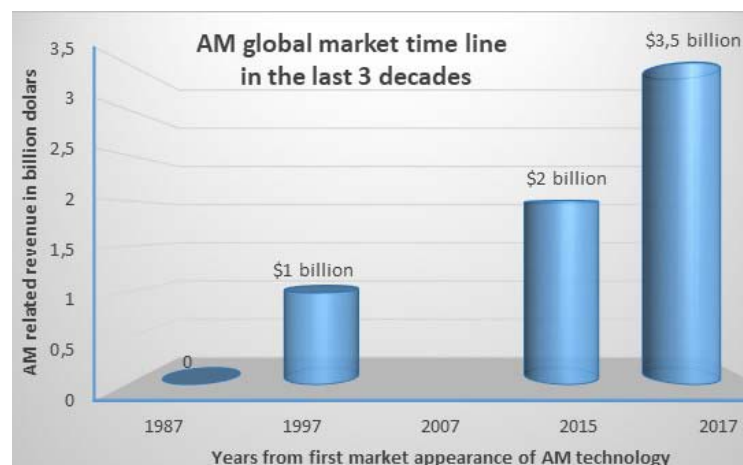


Figure 2. Plots of the growth trends of the additive manufacturing industry.

4. Technology road map and fabrication guideline standards for AM

Surplus availability of materials for AM and their compatibility are among the chronic problems of the field to explore more of its application areas. Moreover, the challenges rise due to AM systems that are designed and developed for particular materials. Thus, innovations on new materials will represent next significant progress expected for the engagement of future researches to tackle the challenges through optimized and different processing conditions. For example, FDM is a plastic based technology, which is usually unfit for those products that require high strength, magnetic and electrical conductivity, application for human implants, etc. In addition, more focus is needed to develop commercial standards for testing of such materials and their resulting parts. Their consistencies in terms of mechanical, physical and metallurgical properties are not yet established for those based on alternative materials. In order to tackle such critical issues, it is important to define roadmaps [27] that facilitate, among others, adoption of industrial standards and effective means of promoting how new material developments and other research outcomes are shared.

There are many additive manufacturing methods and system operation guidelines provided by different standardization companies that depict the methods and specific constraints of machines and listed considerations that provide a set of 'design specifications'. For instance, a committee that develops AM-related standards has been established by the International Organization for Standardization (ISO/TC 261) and the American Society for Testing and Materials (ASTM F42) [28]. This committee has produced standards related to nomenclatures, single processes, flow of processes including required software and hardware, test procedures, quality parameters, means of customer-supplier agreements, and basic elements. Recent additions address data processing and consider the relevance and specify variations to existing standards [20].

As additive manufacturing of metals, polymers, and other materials are entering into a widespread industrial use, it is critical that the infrastructures with additive manufactured parts do not reduce the safety or reliability of the systems. The potential roadmaps are thus expected to indicate the work that is needed to assure safe application of the technology and to identify the unique activities to be performed [29]. In the area of functionally graded materials, in particular, AM technology is considered to provide exclusive opportunities that other conventional methods cannot.

5. Observed research gaps

Despite the fact that AM systems are proved useful in hard-to-reach locations, such as military bases, the international space station, and also widely employed in the medical applications especially for patient-specific orthopedic implants, there is still a need to fill the gap for many consumer products. Furthermore, many more good parts of this technology are yet to come for the well-being of human needs.

Among others, an intensive materials research and development is needed in order to broaden the selection of suitable materials, prepare a database of the mechanical properties of parts fabricated by AM, and determine the interaction between materials and process parameters. The mechanical property of the fabricated parts is still not consistent and the process lacks repeatability of achieved accuracy and surface quality within each machine and across different machines operating under the same principle. Both the applicable design procedures and process modelling and control are considered as critical issues that require closer study in the future research [21].

After rigorous review of recent and on-going researches, the following research directions were identified as some of possible research gaps.

- Design of new alloys using mixed powder solutions to create novel control of microstructures.
- Fabrication of the so-called high entropy alloys, which constitute the latest discovery in design of metallic materials.
- Design of novel material combinations and metal powders to create new functional properties.
- Development of biomaterials that serve as cell delivery media or as biomolecules in the bioprinting field.
- Conducting tests on how mixed powder solutions using additives can be applied to tune solidification and hence microstructural development during the AM process.

- Research on correlation between process parameters, for instance, how the various process parameters affect the material and the resulting component.
- Reproducibility issues, i.e. how to produce the same part with maintained material properties repeatedly in different machines.
- Methodology to obtain minimum iteration of the process.

6. Conclusions

Nowadays, AM techniques are in a position to secure a key role in the modern manufacturing sector to provide 3D part production with different materials composition and this can be a unique turning point of the manufacturing industry. The possibility of producing three dimensional real objects directly from a 3D model data that can have controllable properties will for sure open opportunities for additional research as well as developments for upgrading and implementations of AM. Beyond all the unlimited opportunities to design and produce parts from composite materials as per the client interest and engineering flexibility for customization of products through AM system are the fundamental uniqueness of this technology.

In this article, the opportunities offered by additive fabrication of polymer composites and the accompanied challenges are reviewed and elaborated. Through the process of this study, it is observed and concluded that polymer composite based AM fabrication is at flourishing stages to pave the way to implement 3D/AM system from limited application of prototyping techniques to automated fabrication systems and such needs are not too far away to knock out the traditional manufacturing process soon. Time efficiency and customer satisfaction are the basic parameters fulfilled by AM/3D for demand and supply of products at reasonable costs. The unique characteristics of the technology such as providing excellent adaptation, fabrication with composite materials, i.e. multi-material fabrication of functional customer needs and fabrication of 3D part products with full satisfaction of intended material properties are significant steps in the manufacturing sector. Moreover, the need for complicated parts in diverse sectors such as the automotive, aerospace, biomedical, electronics, robotics industries, as well as fast fabrication of components for maintenance purposes has made high demand of polymer composite product through AM to achieve and provide timely service. Fiber reinforced polymer composite materials, in particular can maximize the near future needs of the upcoming aerospace and automotive industries.

In conclusion, parallel with the necessary advances in the fabrication process technology, broadening its application areas and availability of materials for diverse application, significant work is needed to get acceptance of the technology in the industry. This needs diverse forms of changing the existing mind-set to gain industrial acceptance.

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