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A review of FDM and graphene-based polymer composite

To cite this article: J Mogan *et al* 2021 *IOP Conf. Ser.: Mater. Sci. Eng.* **1078** 012032

A review of FDM and graphene-based polymer composite

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Abstract. Graphene is a carbon that has a unique structure that is excellent in enhancing mechanical, electrical and thermal properties. The fused deposition modelling (FDM) process is a widely used 3D printing method for its low investment and operating cost. Although the FDM process is cheaper and affordable, yet the printed parts are more fragile compare to other 3D printing methods. This paper covers about FDM process and the type of base materials and filler materials. However, the focus is mainly on ABS and graphene. The mechanical properties of ABS/Graphene polymer composite and application of ABS and graphene in the industry were also discussed. Hence, it proved that graphene enhances the properties of ABS. This study is done to improve polymer-based filaments for future references.

Keywords: Acrylonitrile Butadiene Styrene (ABS); Fused Deposition Modelling (FDM); Graphene; Additive Manufacturing; Mechanical Properties; Applications.

1. Introduction

In the 1980s, computer-aided design (CAD) was first introduced by rapid prototyping and this technology specifically provided the realization of user design without applying a mould or other machining preparation. This technology has since been developed exponentially, and in combination with various implementing materials, different forms of technology have been invented and developed. Due to the range of technologies available, additive manufacturing (AM) is referred to more frequently than rapid prototyping. AM has been applied for years, but this method has drawbacks, admittedly, and brings to the world a new age of manufacturing. [1]

Additive Manufacturing (AM) which also known as 3D printing is one of the techniques of adding material to build products from 3D model data, commonly layer after layer built through computer-aided design (CAD), as related to multiplying technology aspect [2]. The material is however melted (fused), cooled and also solidified in this process, fulfilling 3D geometries while not adopting complex models. AM technology is generally referred to as 3D printing [3]. Numerous AM methods depend on plastic lamination, such as laminated object manufacturing (LOM)[4], Stereolithography apparatus (SLA), which rely on the polymerization of images. The production of fused filaments (FFF) and selective laser sintering (SLS) depended on the plastic filaments being melted[5]. Due to the variation of cheap filaments and minimal printing device prices, the FDM technique is more popular than those in industrial applications. FDM 3D printers are commonly used in many such as manufacturing, automobile, medicine and aeronautics for rapid prototyping [6].

One of the most commonly used additive manufacturing techniques is fused filament manufacturing (FFF), which is mainly known as fused deposition modelling (FDM). In reality, the term 3D industries printing is generally used to refer to this technology[7, 8]. The most frequently used thermoplastic polymers are polylactic acid (PLA) and acrylonitrile-butadiene-styrene (ABS), as well as polycarbonate (PC) and polycaprolactone (PCL)[9]. The schematic diagram of the phase of FDM is shown in Figure 1. The FDM raw material will be in the form of a filament which is also heated in the liquefier head to a semi-solid state and then extruded to form a coating on the previously accumulated layer through the heated nozzle. The newly accumulated layer is often solidified due to the dissipation relation between layers retained to the previously accumulated layers[10].While Fused Deposition Modelling (FDM) is also well recognized mostly for operating costs and affordable investment costs, FDM's printed parts are more brittle compared to other typical plastic manufacturing processes, including moulding, injection, CNC, extrusion and blowing[11].

Although FDM produces materials of a reasonably machinable quality, today the world needs much good 3D printed materials than traditional techniques, and they can be obtained by using conventional blends of filament and carbon. Material durability and stability are the main factors in evaluating filament products for the manufacture of finished products using 3D printing. [12, 13]. Graphene nanoplatelets have recently been investigated as possible reinforcement fillers for polymers. Graphene nanoplatelets are tiny particles made up of sheets of graphene. This nanofiller can be used as a multifunctional reinforcement because it has excellent mechanical, thermal and electrical properties with a 2D graphene deformed structure. Therefore, thermoplastic nanocomposite packed with graphene greatly improves mechanical properties and thermal stability. [14-16]. This review paper aims to discuss graphene based polymer composite using FDM process. However, a particular focus will be given to ABS and graphene.

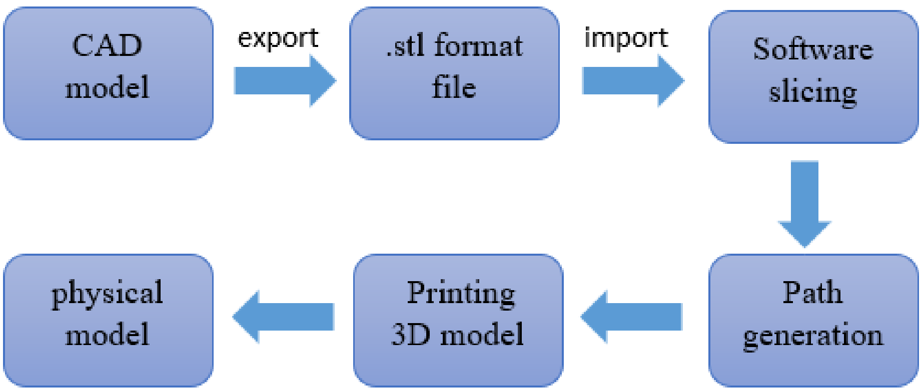

Figure 1. Basic steps for FDM process.

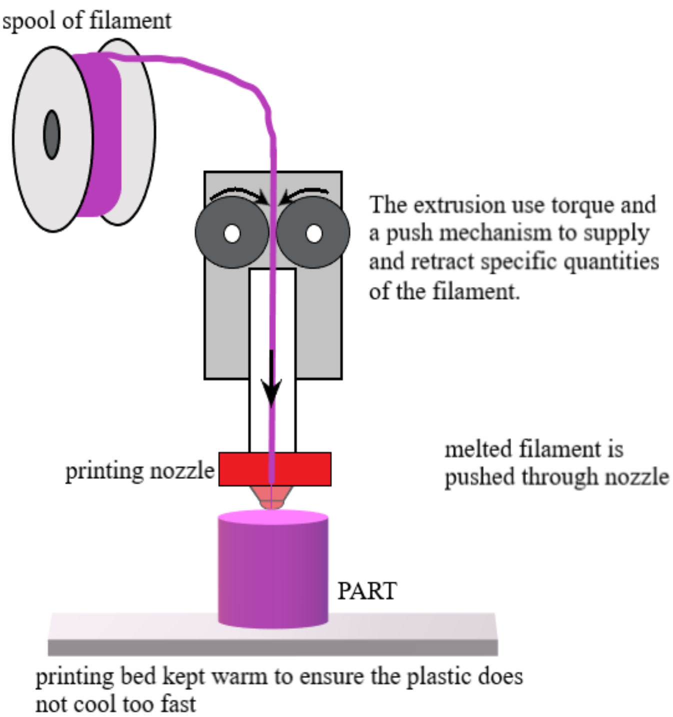

Figure 2. Schematic diagram of FDM process.

2. Type of material used

2.1 Base materials used for FDM process
Acrylonitrile, butadiene, and styrene are derived as ABS. Acrylonitrile is also a synthetic monomer formed from ammonia and propylene. Butadiene is a petroleum hydrocarbon obtained from the steam cracking C4 fraction and styrene monomer is produced using ethyl-a hydrocarbon obtained in the benzene and ethylene reaction dehydrogenation [17].

In general, ABS, which stands for Acrylonitrile Butadiene Styrene, is a thermoplastic that usually functions as a copolymer. The behaviour of thermoplastic materials includes physical, chemical, mechanical, thermal and morphological characteristics such as fracture resistive ability, amorphous vs crystalline, the temperature of melting and glass transformation, surface behaviour, length of the carbon chain, molecular structure, and molecular weight vs molecular density [18].

ABS comprises a system of slightly cross-linked polybutadiene (rubber) to increase the durability of its dispersion, containing styrene-acrylonitrile (SAN). The SAN ratios consist of polybutadiene in the blend and styrene and acrylonitrile in the SAN, the sum of inserted SAN, the size of rubber particles and the distribution of size, as well as the density of rubber cross-links [19]. ABS compositions are 15% to 35% acrylonitrile, 5% to 30% butadiene and 40% to 60% styrene, primarily used in household commercial products as well as Lego brick materials. [20].

Due to its reasonably good workability and outstanding mechanical efficiency, acrylonitrile-butadiene-styrene (ABS) is also commonly used in various electronic devices, household appliances, medical equipment, and others [21]. The ABS content is also known to be cheap, amorphous and manufactured at relatively low temperatures in terms of efficiency [22].

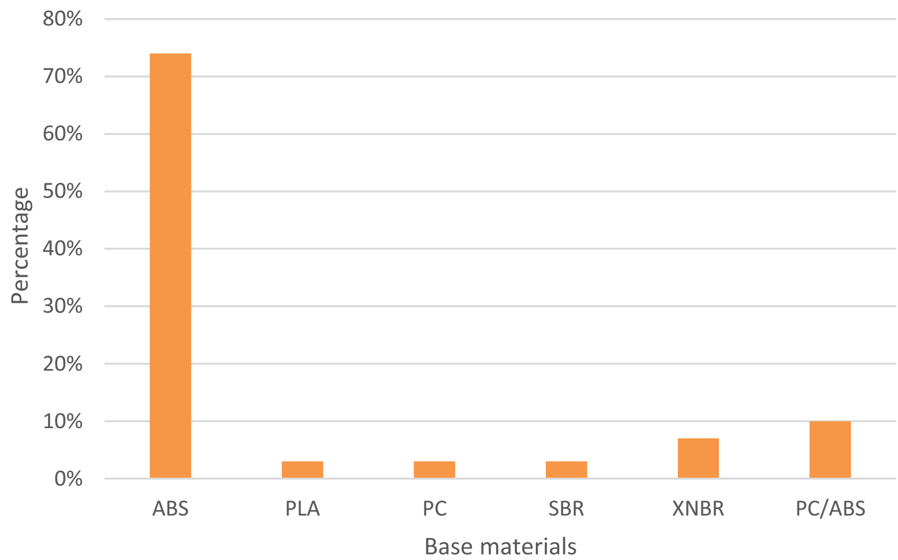

Figure 3. Type of polymers used as base materials.

Figure 3 shows the base materials that have been used. The base materials are Acrylonitrile Butadiene Styrene (ABS), Polylactic acid which is also known as polylactide (PLA), polycarbonate (PC), styrene-butadiene rubber (SBR), Carboxylate Nitrile (XNBR) and PC/ABS. From the figure above, ABS is the most used thermoplastic as the base materials. ABS plastic merges the strength and rigidity of styrene and acrylonitrile with the strength of rubber polybutadiene. ABS is superior for the hardness, shine, durability and electrical insulation properties, although the cost of manufacturing ABS is around 2 times the cost of producing polystyrene[17]. These ABS materials have been widely researched with different agents such as graphene, hollow glass bead filled, nano silica, poly methyl methacrylate (PMMA) and wood plastic to improve their strength.

Table 1. Physical properties of ABS.

Classification	Value	Reference
Thermal Conductivity(K)	0.1 to 0.15 W/m-K	[17, 23]
Density(ρ)	0.9 g/cm3 to 1.53 g/cm3	[23, 24]
Specific Gravity	1030 kg/m3	[25]
Tensile modulus	2 to 2.4 GPa	[23, 25]
Melting Point	230 to 255 °C	[25]
Glass temperature	110 °C	[23]
Solubility in Water	Insoluble	

2.2 Filler materials used for FDM process

Graphene is an atomic carbon layer with minimal thickness and unique electronic structures, resulting in excellent mechanical and electrical properties[26]. It is also powerful, almost transparent, lightweight, and has good heat conductivity and electricity. Carbon nanotubes (CNTs) and graphene are both excellent materials for applications in electrochemistry. Compared to CNTs, graphene has gained popularity due to its economic ease and greater purity[27]. The use of constantly dispersed nanocomposites is motivated by the use of materials with improved mechanical properties required for different applications[28].

Graphene is a two-dimensional hexagonal carbon atom lattice and the fundamental building block for other allotropic carbon types, including nanotubes of carbon, fullerenes, graphite and diamonds. Graphene's remarkable properties are linked to the electronic structure and carbon atoms arranged in the hexagonal planar lattice. Therefore, graphene is an ideal reinforcement material to improve thermoelectrical efficiency and structure of the metal matrix composites[29].

Graphene has a two-dimensional geometric feature that can deliver very high aspect ratios and is responsible for the highest value over its surface-to-volume proportion. As the suitable candidate for a reinforcing material in polymer nanocomposites, its significant price of Young's Modulus and tensile strength enhances its mechanical properties. The mechanical strengths of polymer nanocomposites are affected by the huge increase of graphene. It not only does its incorporation as nanofiller transform the heat distortion temperature, hardness, stiffness, toughness, and also economic ease due to the unique nanostructures. This is subject to the aspect ratios and large surface-volume also polymer matrix connection[26].

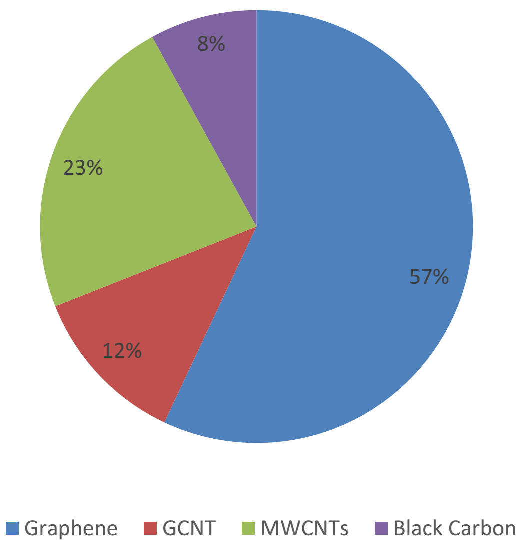

Figure 4. Type of carbon used as filler materials.

Figure 4 shows the type of carbon used as filler materials. The filler materials are graphene, graphene oxide-carbon nanotubes (GCNT), multiwalled carbon nanotubes (MWCNTs) and black carbon. Based on the studies conducted, graphene is the most used carbon filler compared to other carbon filler. This is because nanocomposites of the polymer network with graphene and its subordinates as fillers have shown exceptional potential for numerous important applications, such as gadgets, environmentally sustainable energy vitality, aviation and car companies. As stated some time recently, 2-D graphene has better electrical, mechanical and warm properties as well as other interesting elements, including a higher proportion of viewpoints and a greater specific surface area compared to various fortifications[30].

Table 2. Physical and mechanical properties for graphene.

Classification	Value	Reference
Density	1.06 g/cm3	[29]
Surface area	2,630 m2 /g	[29]
Thermal conductivity	5300 W/m.K	[29, 31]
Tensile strength	130 GPa	[29]
Young's modulus	1.02 TPa	[29, 31]

3. Mechanical properties of ABS/Graphene

Testing mechanical properties helps to evaluate and design materials and products so that they last longer are more efficient and less expensive. It also helps in designing products that are desired. Several mechanical test studies have been carried out in the literature to study the mechanical properties of polymer composites, such as tension test, compression test, impact test, bending test, hardness, creep and fatigue. Most mechanical properties studies of ABS/Graphene materials manufactured by the FDM were in tension scope in the papers read. There is some researcher has discovered to study impact, bending, flexure and hardness.

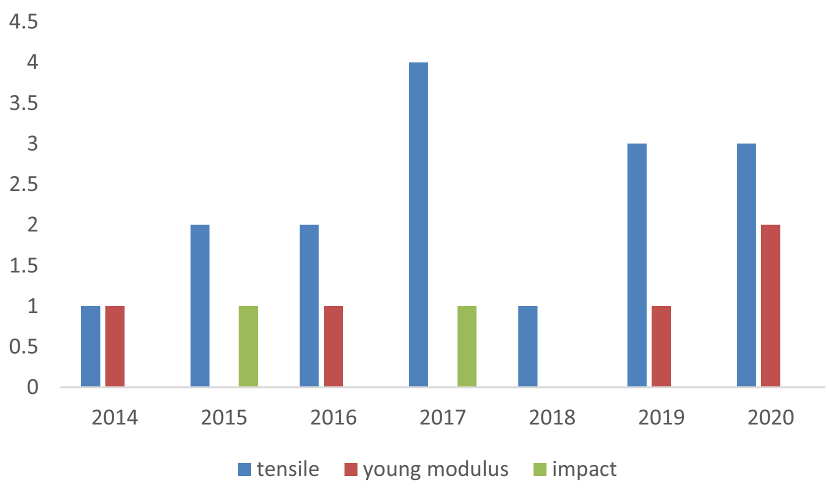

Figure 5. Type of mechanical properties tested from year 2014 to 2020.

Figure 5 shows the mechanical properties being testing from year 2014 to 2020. Based on the study conducted, the most preferred mechanical properties to be tested is the tensile test. Meanwhile, young modulus are second highly tested mechanical properties. Impact test is the least tested mechanical properties. In order to obtain the material parameter that includes ultimate tensile strength, yield stress, young's modulus, toughness and ductility, the tensile test is applied to a sample. In preparing a sample for the test, there are many options for shapes and designs. Even though there are many choices in the design of the test sample, those designs must follow the design of the ASTM standards.

One of the common designs used by most researchers was a flat dog-boned shape sample, [32]. Different forms, such as round specimens, were also used in tensile studies, according to ISO-7500/1 [33], CT specimens (ASTM E 647-08) [34], and non-standard shapes such as struts [35], and real part shapes [36].

Many researchers have stated that graphene has enhanced the mechanical properties of ABS. Hence, it is proved that ABS/graphene polymer composite material has better mechanical properties compared to ABS based materials. Sithiprumnea (2016) stated that samples are produced through different processes such as extrusion, compression moulding and also fused-deposition modelling, the thermo-mechanical properties of ABS and also its nanocomposites were compared. The existence of graphene nanoplatelets has increased the ABS tensile modulus[9].

The use of graphene nanoplatelets in ABS indeed increases ABS conductivity. The disadvantage is that the polymer matrix structure has been interrupted, thus reducing its impact strength, but the polymer elasticity somehow has increased. The images of morphology display strong dispersion and close interaction of the graphene platelet filler and polymer matrix, which has a positive effect on the electrical and mechanical properties of nanocomposites[37]. The fabrication of ABS/graphene composites will also potentially expand the applications and capabilities of 3D printed products.

4. Application of ABS and Graphene in industries

Acrylonitrile Butadiene Styrene (ABS) is a thermoplastic resin commonly used because of its excellent mechanical and processing properties in industrial sectors such as automotive, electronics, machinery, telecommunication instruments and construction [38, 39]. In addition, ABS is chosen as the manufacturing material in the automotive industry where it is utilized to manufacture bumpers and pillar trims[40].

Due to their outstanding electrical, mechanical and thermal properties, carbon nanofillers (carbon nanotubes, graphene oxides, carbon nanofibers, and reduction of graphene oxide) have been proved to be of great involvement in polymer composites. The relatively attractive materials used for different applications such as transparent electrodes, field-effect transistors, optoelectronics, energy storage materials, composites, and biosensors have been identified to be carbon nanofillers[41]. Besides that, graphene and carbon nanotubes (CNTs) are also amazing materials to be used in electrochemistry. Compared to CNTs, graphene has gained popularity due to its greater purity and economic ease[42]. Other than that, In addition, graphene has been used to coat metallic materials, indicating its potential to enhance tribological behaviour and extend the life span of these materials[43].

5. Summary

The types of commonly used thermoplastic and carbon have been analysed in this article. ABS has been used as the base material while the filler material is graphene. FDM printed products are easily breakable compare to other manufacturing processes due to their printing orientation and also material. Hence, based on the studies that have been conducted, graphene plays a significant role as a filler material in enhancing the mechanical properties of ABS. In general, based on the research that has been performed, the most commonly used mechanical properties to measure ABS/Graphene samples is tensile strength. The common application of graphene is in the electronic industry such as biosensors. Other than that, ABS has also been used in various industries where it is utilized to produce telecommunication instruments and electronic devices. Thus, due to its high compatiblity characteristics, different forms of graphene are being incorporated as electrodes, field-effect transistors, energy storage materials, and composites. Therefore, some research studies suggested implementing graphene as a filler in order to enhance the properties of polymers.

Acknowledgments
The authors gratefully acknowledge the support provided to the facilities and resources for this research by Universiti Malaysia Pahang (www.ump.edu.my). The authors would like to thank their support for the internal grants of Universiti Malaysia Pahang (RDU1903134, RDU190354) and the support is given by the Ministry of Higher Education, Malaysia, to the Fundamental Research Grant Scheme (FRGS.) FRGS/1/2019/TK03/UMP/02/15.

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