



# Potentials and challenges of additive manufacturing techniques in the fabrication of polymer composites

Smith Salifu<sup>1</sup> · Olugbenga Ogunbiyi<sup>2</sup> · Peter Apata Olubambi<sup>1</sup>

Received: 13 June 2022 / Accepted: 12 August 2022 / Published online: 18 August 2022  
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## Abstract

As a promising technology capable of transforming the conventional manufacturing techniques, the use of additive manufacturing (AM) has span beyond the prototyping it was initially known for, and its use is currently revolutionising the future of the manufacturing and research world. A review of some of the advances made in the additive manufacturing of polymers and their composites is presented in this paper. Some of the advantages and disadvantages of the different AM techniques used in polymer composites (PC) fabrications are presented, and the different areas of applications of the AM fabricated PC are highlighted. Also highlighted are some of the potentials and challenges associated with the fabrication of components using 4D printing. Finally, the paper presents the prospects and the endless opportunities that abound with the AM of polymeric materials.

**Keywords** Polymer composites · Fibre · Matrix · Reinforcement · 3D printing · 4D printing

## 1 Introduction

Over the years, the terms additive manufacturing (AM) and 3-dimensional (3D) printing have been used interchangeably, and the technology is currently at the forefront in the innovation of materials, design and engineering, production waste management, cost reduction, and increasing efficiency. It is no more news that this technology has come to reshape the manufacturing processes, and it is worth noting that within its short period of AM existence, its industrial, economical and societal impacts have been tremendous [1]. Historically, the advent of AM technology dates back to the 1980s when its application was limited to prototyping or manufacturing of small products. Depicted in Fig. 1 is a chart showing the advancement of AM technology from its advent to date.

From the year 2009, AM technology started experiencing rapid development due to the introduction of new dimensions in engineering applications that soothes the different

industrial applications. Figure 2 shows the yearly research publications in additive manufacturing, and Fig. 3 shows the geographical distribution of patents in the field of study. Although the use of AM technology is still relatively new and currently at its young stage, a lot is still being done to develop high, quality complex components from different materials and multimaterials with high precision and performance level.

The flexibility provided by polymer composites in terms of the properties of the final product, the several raw material options, design, and the continuous improvement in techniques used in their synthesis [2, 3], coupled with their cost-effectiveness and availability in commercial quantity as compared to other materials such as ceramics and metals for use in a low-cost application makes it an area of research interest [4]. Polymer composites are gradually becoming the prime choice of materials for critical applications such as automotive, medical devices and aerospace, because of their tailored and enhanced material properties [5]. Compared to conventional materials, polymer-based nanocomposites avail a greater superiority margin because of their higher strength-to-weight ratio, flexible manufacturing process, ease of fabricating products with customised properties, and their high corrosion resistance [2].

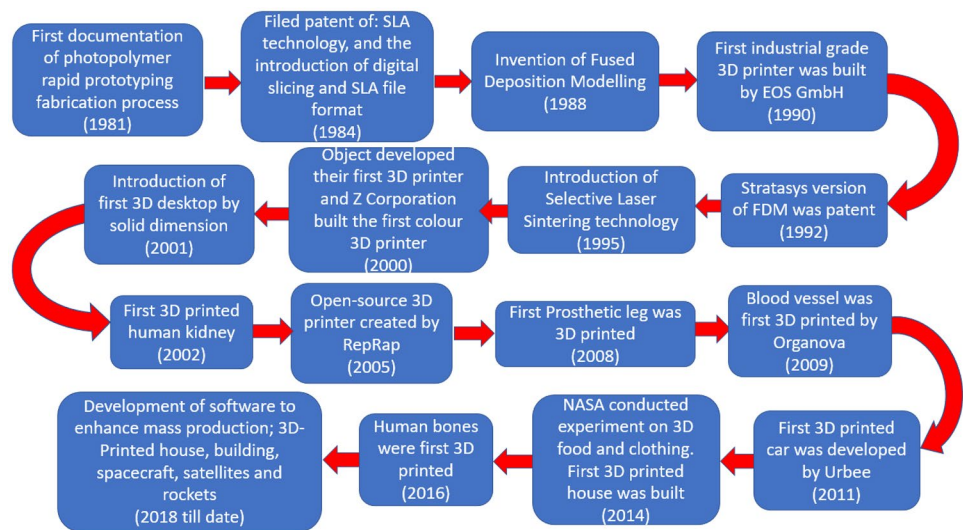
Generally, AM involves the building of a 3-dimensional (3D) product from a computer-aided design (CAD) generated

✉ Smith Salifu  
smithsalifu@gmail.com

<sup>1</sup> Centre for Nanomechanics and Tribocorrosion, School of Mining, Metallurgy and Chemical Engineering, University of Johannesburg, Johannesburg, South Africa

<sup>2</sup> Department of Mechanical and Mechatronics Engineering, Tshwane University of Technology, Pretoria, South Africa

**Fig. 1** Chart showing remarkable achievements in additive manufacturing from its advent in the 1980s until date



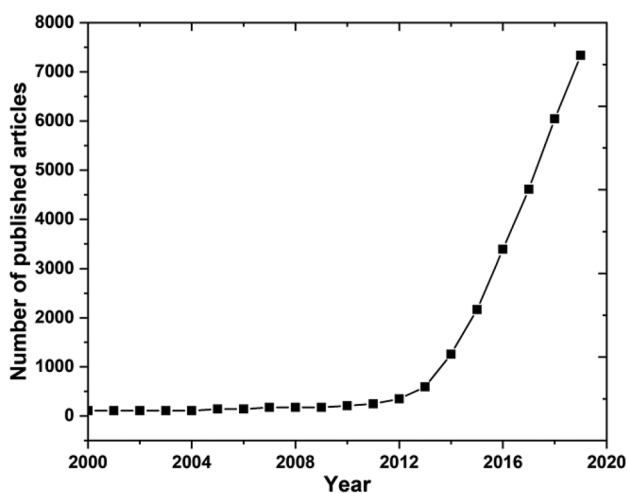
model that gives unprecedented room for the digitisation of the manufacturing sector. Leveraging the various technological platforms and software available, computational speed and connectivity, the possibility of achieving appreciable success with this relatively new manufacturing technology is limitless. Also, the available technologies have created room for improvement in design accuracy, reliability, and the possibility to manufacture customised products with limitless design flexibilities.

The automotive industry, aerospace, electronics, consumer goods, and biomedical fields are some of the areas of applications that have benefited from the use of AM technology [2]. With the discovery of possible improvement of mechanical performance of fibre-reinforced additively manufactured or 3D printed plastic/resin products, AM of polymer composites has experienced tremendous expansion. The

use of this technology has enjoyed exponential patronage, particularly in the development of components used in novel applications, and the development of innovative ways of 3D printing materials in an efficient and cost-productive way.

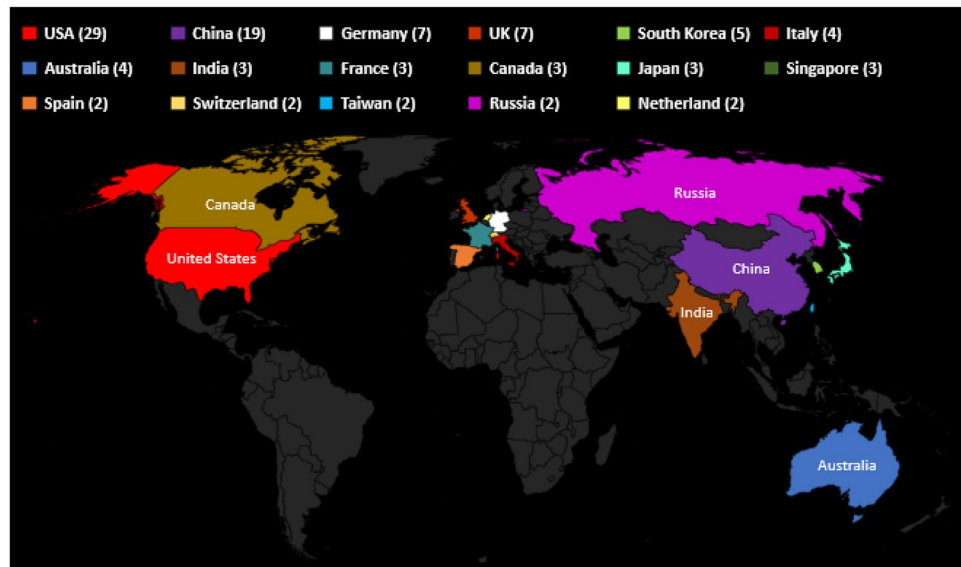
The performance improvement of medical components, like tailored prostheses (a component designed to replace missing body parts or make the parts work properly) achieved through the provision of unique and innovative designs that uses hydrogels and different advanced materials to enhance the final product's quality and its biocompatibility, is a major area of focus of AM technology. With the current emergence of 4D printing, the ease of forming complex 3D structures which are capable of assuming different predetermined shapes and forms, when exposed to different levels of environmental stimuli, is on the increase. Also, the use of programmable smart materials, i.e. stimuli-responsive polymers, and the cumulative effect of shape programming are responsible for the unique shape mechanism exhibited by the process. As an evolving technology, AM can proffer solutions to digitally manufactured products without a complicated framework and human intervention, and the use of the technology seems limitless.

This review gives a concise account of the progress and the state-of-art additive manufacturing techniques, with emphasis on polymeric materials such as polymer composites, thermoplastic polymers, nanocomposites, biopolymers and fibre-reinforced plastics. Nevertheless, AM of components using metals, multi-metals and ceramics are discussed when required, for better understanding. Also presented in this chapter is the stride made by 4D printing of stimuli-responsive polymer materials during the production of functional biomaterials used for medical purposes. In an attempt to present the potentials of AM technique in polymer composites, the different methods and materials used, processes developed and their industrial applications are



**Fig. 2** Annual additive manufacture research article publications indexed in the Scopus database from the years 2000 to 2019

**Fig. 3** Geographical patents distribution on additive manufacturing based on the Scopus database from the years 2000 to 2019



presented. Lastly, the challenges faced with the use of the technology (AM) and its prospect with polymer composite are highlighted.

## 2 Additive manufacturing processes

Fundamentally, there is a difference between the traditional manufacturing and AM process as the component manufactured using AM technology is built via a layer-by-layer method [3–5]. This approach made the use of AM technology a preferred choice for most industries that use a wide range of materials such as ceramics, metals, polymeric materials (in the form of composites), hybrids and functionally graded materials, and hydrogels (in the form of liquid), solid gels and viscoelastic. Based on the flexibility of AM process, its classification is very broad; thus, AM process can be classified in different contexts as depicted in Fig. 4. The figure shows the different perspectives of AM classification. Based on the report from ASTM (ASTM F2792-12a) and the classification processes shown below, there exist more than 50 different AM technologies [6]. As such, a set of standards was formulated by ASTM that classifies AM processes into seven general categories (ISO/ASTM 52,900:2015).

In accordance with the formulation methodology of the final component, ASTM classifies AM process into seven types namely [7]:

- Jetting
- Binding jetting
- Powder bed fusion
- Vat photo-polymerisation
- Energy deposition
- Material extrusion and

- Sheet lamination

With respect to the base material processing medium, AM can be classified as ultraviolet rays, thermal means, laser beam etc., and parameters like part strength, speed of fabrication, resolution, cost, the volume of built, surface finish and quality are used for the evaluation of these processes. These processing parameters are evolving continuously, thus making it possible to economically fabricate a bigger and more complex product in a more flexible way [7–15]. Of all these processing parameters mentioned, resolution and fabrication speed are the most important because they greatly influence the outcome of the final products produced. With respect to AM process methodology used in the formation of products, the evaluation is usually done base on the fabrication speed and resolution. In view of this, several 3D printing systems have been developed and successfully used in the production of several advanced and sophisticated structures for industrial and research applications. The classifications of AM according to ASTM and the advantages and disadvantages of the specific AM processes used are briefly discussed in the subsequent section.

### 2.1 Material jetting

Known as one of the most accurate and fastest AM or 3D printing processes, the material jetting process requires that the support materials and the build of the liquid material droplets are jetted selectively on the build platform in such a way that the layers previously deposited are partly softened by the new droplets. Thereafter, the deposited layers are cured/solidified with ultraviolet light into a single piece, where they can be removed or separated from the platform. The process of material jetting can be akin to a 2D inkjet

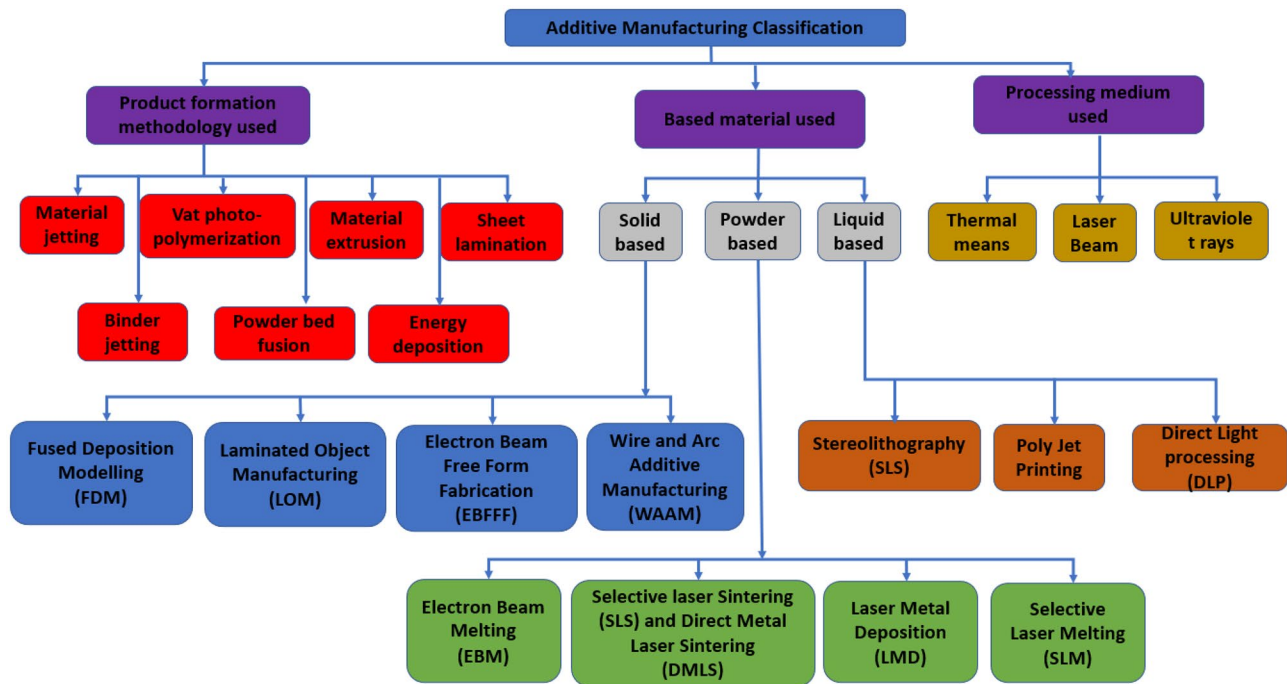


Fig. 4 Different contexts for the classification of additive manufacturing process

printer [16, 17], and the polymer employed for the process is usually thermoset photopolymers that are usually available in liquid form (i.e. acrylics). Other fully transparent rubber-like and acrylonitrile butadiene styrene (ABS)-like materials are commercially available for use with this AM process. One of the key strengths of this process is its multimaterial printing ability, which makes the process suitable for the creation of realistic haptic and visual prototypes that possess smooth surfaces similar to those created using injection moulding (possessing homogeneous thermal and mechanical properties). Despite the wonderful advantages the use of this process offers, it has some limitations such as:

- Poor mechanical properties
- A high material cost which limits its usage in some applications
- Photosensitivity
- Mechanical properties degradation with time

Based on these limitations, components produced using the material jetting process are predominantly used for non-functional prototypes [18].

An example of material jetting process is the polyjet printing process that uses inkjet technology in the creation of 3D parts. A typical inkjet head consists of the photo-resin that deposits photo-resin based on the CAD file as it moves along the X and Y axes as depicted in Fig. 5. After the deposition is completed, an ultraviolet lamp is used to

cure the deposited photo-resin layers until the desired part is completely created [19].

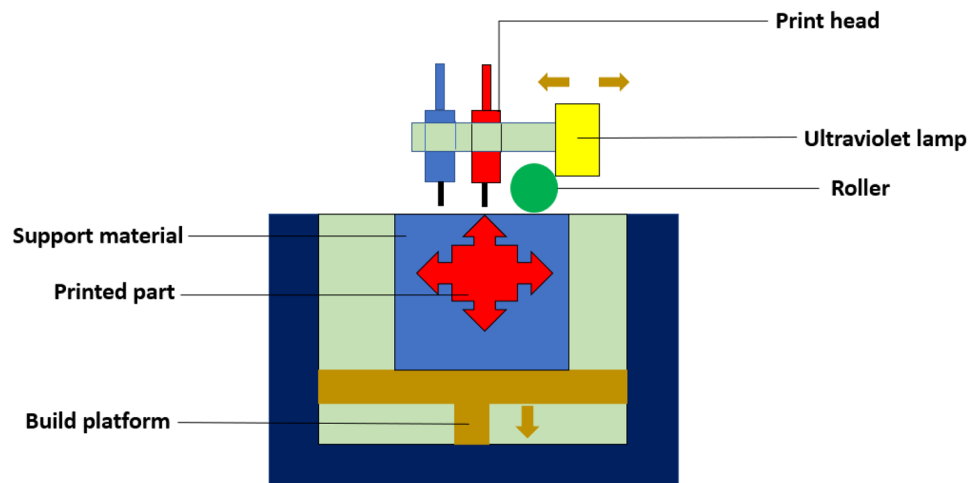
## 2.2 Binder jetting

In a binder jetting AM technique, CAD deposited liquid bonding agents are used in joining the powder particles layers that are selectively deposited on the build platform to form the required part. The printer head in the binder jetting technique is designed to easily drop the bonding liquid onto the selected powdered particles on the platform. As soon as a layer is formed, the platform moves down to allow for the formation of the next layer. The main advantages of the binder jetting technique include:

- Allows for the creation of complex designs
- Higher printing speed and
- Support structure freedom

The other advantage of the blind jetting technique is the fact that it allows the use of a different variety of materials such as metals, polymers, ceramics and sands of different colours, and also, gives room for manufacturing large and complex components at a relatively lower cost, as compared to many direct 3D printing processes. The ability to manufacture high-value products using structurally robust materials is another aspect where the blind jetting process displays its uniqueness. In spite of the significant success recorded

**Fig. 5** A typical inkjet head with photo-resin



in the use of this AM technique or process, there is a need to conduct extensive research on the use of the technique in order to generate fundamental data that are needed for the large-scale robust implementation of the technology [20–24].

### 2.3 Vat photo-polymerisation

Vat photo-polymerisation is an AM technique that uses liquid photo-curable resin vat in conjunction with a suitable laser to construct solid products whose photosensitive liquid is selectively hardened layer by layer into a 3D solid. The cured part ascends from or descends into the vat of the photosensitive liquid resin. The use of this manufacturing technique allows for rapid prototyping and manufacturing of parts with very high resolution, and an exceptional surface finish. Comparatively, this technique is quite expensive and the brittleness associated with the produced product over time is a major concern. Despite these shortcomings, this AM technique has proven to be effective in model concept creation, rapid prototypes creation and the creation of complex geometrical parts. Generally, the height of the layer in the Z-axis that is often used in the 3D printer resolution definition can be adjusted between 25 and 300 microns on the recently developed vat photo-polymerisation printers for a better print. An example of a modern printer in this category is the Formlabs stereolithography (SLA) 3D printer, whose operation requires a compromise between quality and speed [25–27]. Compact and highly sophisticated SLA 3D printers are being developed due to the advancement in technology and the innovative use of the formulations of SLA resin with different ranges of mechanical, optical, and thermal properties to meet the standard industrial and engineering thermoplastics. This advancement has created room for an increase in innovation and the introduction of technology that supports a wide range of industrial businesses such as

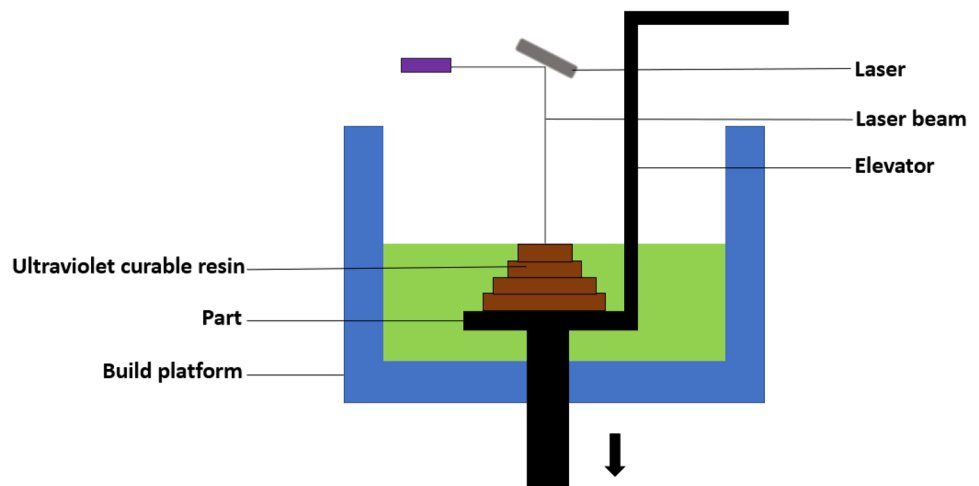
manufacturing, engineering, education, jewellery, audiology, and healthcare [28].

### 2.4 Stereolithography

Known as the first AM process to be developed, stereolithography (SLA) process forms its parts through the exposure of a photosensitive resin to an ultraviolet laser as depicted in Fig. 6.

Upon the exposure of the photosensitive resin to the ultraviolet laser, a solidified part layer is formed, and more layers continue to form as the resin is continuously exposed to the resin by lowering the build platform as specified in the CAD data. It is worth noting that a diverse material range can be used in conjunction with the SLA technique in the fabrication of products. A typical example was the fabrication of ceramic composite parts from an ultraviolet curable  $ZrO_2 - Al_2O_3$  ceramic composite pastes using the SLA technique before debinding and sintering the created part [29]. Employing this AM technique successfully, novel biocompatible photochemistry was used to create personalised medicine geometries and drugs that consist of ascorbic acid encapsulated in polyethylene glycol dimethacrylate-based polymer network, which was polymerised using riboflavin as the photoinitiator [30]. The success recorded in the use of novel biocompatible photochemistry and 3D printing confirmed that ascorbic acid can be loaded and released successfully as a model agent. This finding has opened up a new kind of manufacturing procedure capable of encapsulating ascorbic acid, and other water-soluble vitamins suitable for drug delivery in the pharmaceutical industry [1]. Also, passive stabilisers have been successfully used to support the 3D printing board of SLA on moving vessels on the sea, and the success recorded gave birth to the lithographic 3D printing used in naval and voyage designs for oceanographic applications [31].

**Fig. 6** Stereolithography (SLA) process for parts formation



## 2.5 Direct light processing

The direct light processing (DLP) technique is similar to the SLA process. Unlike the SLA process where ultraviolet light is used to cure the photosensitive resin, a DPL projector is used for curing the photosensitive resin as depicted in Fig. 7 [32].

Aside from the use of this process in the fabrication of polymer parts, DPL has found significant relevance in biomedical applications where hydrogel, bioinks and ceramics are used. Worthy of mentioning is the promising result obtained when DLP was used to print scandia-stabilised zirconia ceramic parts [33] and the development of photo-curable chitosan bioink (CHI-MA) printing into complex 3D hydrogel structures with good biocompatibility, high fidelity and resolution [34]. Using this AM technique with bio-composite materials, scaffolds for bone grafting purpose were successfully fabricated [35]. Not too long ago, photosensitive resin-based technology has been employed with the use of  $ZrO_2$  to fabricate ceramic teeth for biological engineering [36], while compatible poly-L-lactic acid (PLLA) resin has been successfully used with DLP to produce hard tissue scaffolds [37], and the results obtained showed that the polymer and the DPL

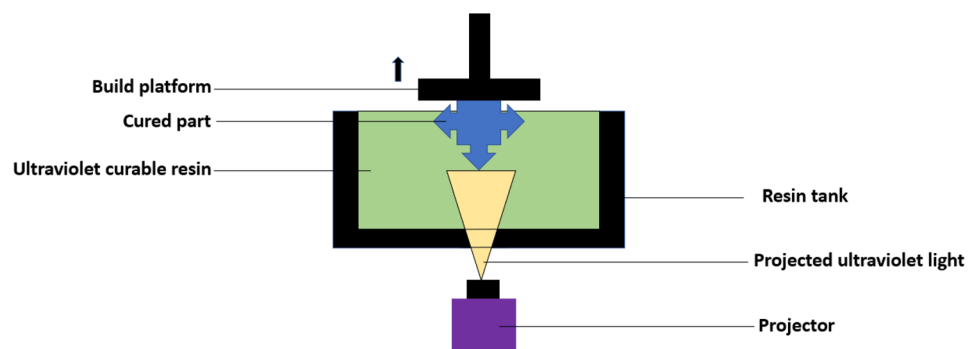
AM method are suitable for the fabrication of scaffolds with complex structures [1]. Also, success has been recorded with the use of the DLP technique in the fabrication of polymer-based microfluid chips [38]. Hence, the DLP technique has displayed exceptional ability in the production of products with evenly distributed cells because of its fast printing speed, and the photo-polymerisation of the polymer.

## 2.6 Powder bed fusion (PBF)

In the powder bed AM process or technique, the powder particles are fused based on the CAD object specification in CAD by using thermal sources such as laser beams. After which, a recoat blade is used to make the formed layer uniform before the formation of the next layer. The PBF process comes in different variants and can be distinguished based on the type of material employed and the heat source used. The common categories of the PBF AM process are:

- Electron beam melting (EBM)
- Selective laser sintering (SLS)
- Selective laser melting (SLM)

**Fig. 7** Direct light processing technique used in curing photosensitive resin



The different PBF additive manufacturing processes come with their merits and demerits, and these strongly depend on the area of application [39]. During the SLS process, sometimes called laser PBF (PBF-LB), powdered polymeric materials like polyetherketoneketone (PEKK) or nylon are sintered in such a way that the 3D parts are created by the successive build-up of powdered material layers over other layers. Using this PBF technique, each successive layer of powder particles is sintered with the aid of a laser beam as depicted in Fig. 8, until the final part is completed.

Some selective laser sintering (SLS) techniques give higher resolution on a microscale and, thus, are referred to as micro selective laser sintering ( $\mu$ -SLS) because they are capable of producing parts with features and size resolution that is less than  $5\mu\text{m}$  [40]. The use of  $\mu$ -SLS is extensive, particularly for the fabrication of actuators on a microscale, micro-optoelectronic components, sensors, etc. Selective laser melting (SLM) on the other hand melts the powder completely with the use of a laser rather than sintering as obtained in the SLS technique. Generally, the SLM is applied to metallic powders such as stainless steel, aluminium alloys, titanium and its alloys. To avoid possible nitriding or oxidation of the consolidated or bulk material, the process is carried out in the presence of an inert atmosphere (i.e. argon), which must be incorporated into the build chamber. The electron beam melting (EBM) process on the other hand is similar to that of SLM but with the laser in the SLM process replaced with an electron gun. Thus, the EBM process is sometimes referred to as the PBF-EB process.

The less popular categories of the PBF techniques are:

- Thermal powder bed fusion
- Fused with agent and energy

The HP's multi-jet fusion falls under the fused with agent and energy category of the PBF technique because the powder bed is heated uniformly at the start, after which the fusing agent allows the bonding of the powder to form 3D

geometrical parts. It has been reported that with the Blue-printer of the Danish Company, 3D parts are being created by thermal fusion through the use of selective heat sintering (SHS) of the thermoplastic powder [41].

Some of the advantages of the PBF process include [1]:

- Rapid prototyping and improved production volume
- Reduction in material wastage
- Reduction in production time
- Ease of recycling unused powder
- Building graded parts with high functionality
- Creating parts with good resolution
- Creation of customised parts due to design flexibility
- Elimination of parts machining
- Ability to increase production by populating the build area with several parts

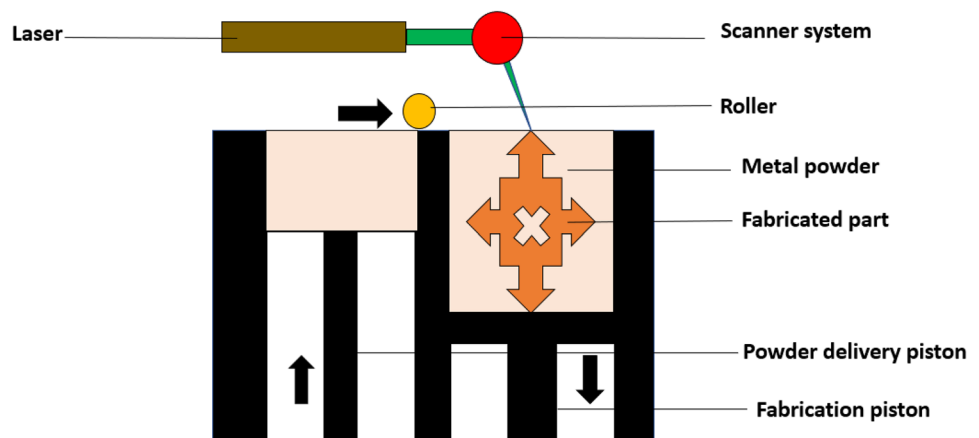
Some of the disadvantages of the PBF process include [1, 19]:

- Slow and long print time
- Post-processing and parts removal are often required for the optimisation of the final properties of the produced parts
- Powder grain size is responsible for the surface quality of created parts
- Increase in cost due to the high quantity of powder usage
- Possibility of thermal distortion
- Weak surface texture/structural properties

## 2.7 Material extrusion (ME)

The AM process, known as material extrusion (ME) and sometimes referred to as fused filament fabrication (FFF), is one of the most used AM processes. In this process, spool of material (usually a polymer) at constant pressure is pushed through a heated nozzle in a stream such that the desired materials are deposited selectively on a layer-by-layer

**Fig. 8** PBF technique for sintering of layer of powder particles with the aid of a laser beam



basis in accordance with the design to create the 3D part as depicted in Fig. 9.

In 1990, the use of FFF technology was improved and commercialised by Stratasys company, and the company adopted a new name, called Fused Deposition Modeling (FDM) for the technique [1]. With the use of this technique (FDM), different materials could be extruded, and thermoplastics such as acrylonitrile butadiene styrene (ABS), acrylonitrile styrene acrylate (ASA), polylactic acid (PLA), high-impact polystyrene (HIPS), polyetherimide, polycarbonate, aliphatic polyamides (PA, Nylon), thermoplastic polyurethane (TPU), and high-performance plastics like polyetherimide (PEI) and polyether ether ketone (PEEK) are the most used materials [7, 42–44]. The FDM technique is been used by different academia, industries and consumers for prototyping and functional components fabrication using both commodity and engineering plastics [45–48]. Employing this AM technique, preformed fibres with uniform material properties and size are fed via the rollers and nozzle, and the extruded filaments solidify as a result of heat loss and then fuse out with the layer formed beneath it. Should a layer be built in a printing cycle, the print head is either lifted or the plate lowered to allow for the next layer to be printed. The flexibility of this technique (shown in Fig. 9) makes it possible for the creation of parts with complex shapes because the technique allows for the extrusion of materials through nozzles with multiple heads, such that one of the nozzles is responsible for part modelling and the other responsible for modelling the support material [45]. Through the use of a post-processing technique such as water jetting, the support

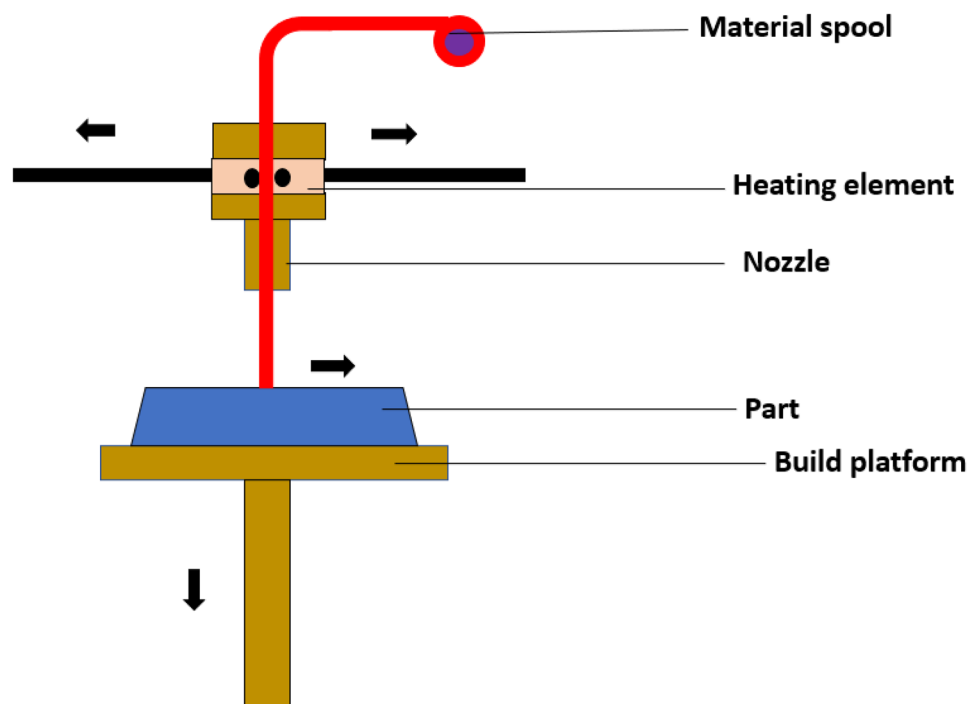
materials can be easily removed upon the completion of the printing. The FDM machine comes in different varieties and, often, differs in properties like resolution, print quality and speed [48]. Some of the FED machines like the Cartesian FDM machine allow the print head to travel or move along the XY plane while the bed is fixed in the X-direction. The Polar FDM machine on the other hand allows the movement of the printer heads in the XYZ direction while the bed is fixed in the Z-direction. In the Delta FDM machine, the print heads are allowed to travel in the printing environment.

Aside from the fabrication of traditional plastic components FDM technique is used for, it has been adapted for use by industries for the development of innovative products such as soft magnets [49], 3D printed microfluid, electrochemical sensing devices, [50], a system for drugs in the pharmaceutical industry and for personalised medicine [51–54], and the design and fabrication of complex porous scaffold for tissue engineering and biomedical applications [55].

The use of the fused deposition modelling AM technique poses the following advantages:

- Significant reduction in the cost and production time for the creation of models/functional prototypes
- End-use parts fabrication without the need for tooling or machining
- Material processing and handling flexibility
- Manufacturing tools fabrication without expensive tooling or machining at a reduced cost
- Absence of unbounded loose powder

**Fig. 9** Fused filament fabrication (FFF) technique for selective layer-by-layer deposition of desired materials





- Faster product marketing
- Solvent removal is not required

In addition to the above advantages, the FDM 3D printing technology is capable of extruding paste-like materials like concrete ceramics and chocolate. The advancement in this technique has led to significant progress in ink material extrusion and the advancement of the liquid deposition modelling (LDM) technique [56].

The progress recorded with the use of the FDM process led to the development of a new technique known as composite filament fabrication (CFF). With the CFF technique, the printer is equipped with multiple extruders which allow the speedy opening of multiple materials and 3D printing of composite materials. In a typical CFF printer with two nozzles, the extrusion of the material is performed by one of the nozzles. The outer shell and matrix formed by the plastic filament are laid by one of the nozzles while a continuous strand of the composite fibre such as Kevlar, fibreglass or carbon is deposited on every layer by the second nozzle. The embedded continuous composite fibre strands inside the 3D printed parts help to improve the strength of the built component [1].

## 2.8 Sheet lamination

In the sheet lamination AM process, stacking and lamination are the used objects, and one of the following methods can be used for the lamination:

- Ultrasonic welding
- Bonding or
- Brazing

The desired final shape of the product can be achieved through CNC machining or laser cutting, while the final product is then made either by.

- First cutting the sheet material to the desired shape and the bond to the previous or initial layer to produce a 3-dimensional geometry or
- Bonding the sheet material layers together before cutting into the desired shape

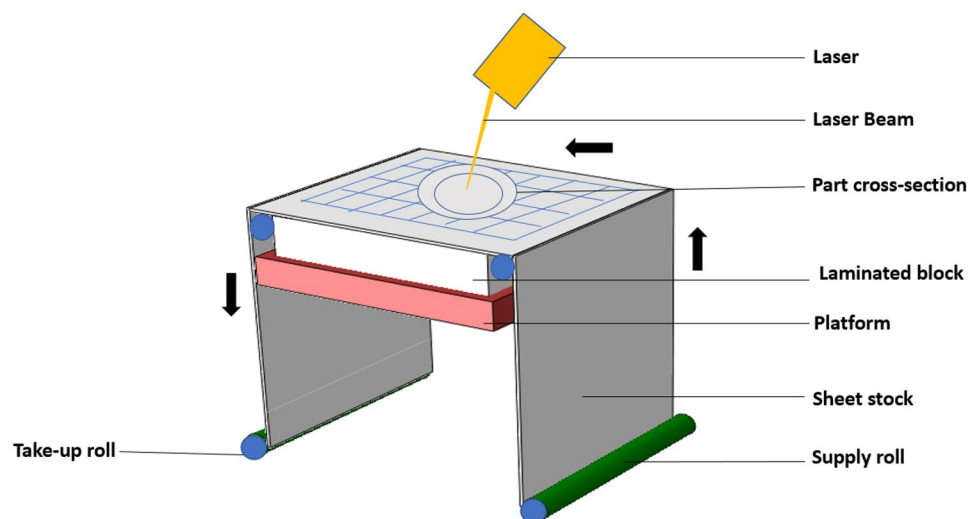
Metal sheets, plastic and paper are some of the materials used in the sheet lamination process, and different sheet lamination techniques are employed for different purposes. Based on the techniques used in bonding the sheets together, sheet lamination can be categorised into:

- Ultrasonic additive manufacturing (UAM)
- Lamination object manufacturing (LOM)
- Plastic sheet lamination (PLS)

Ribbons of metals or sheets that are bonded together through ultrasonic welding are used in the UAM process while in LOM, materials like polymer composite, paper tapes filled with metals, ceramics and paper are used, and they are bonded with adhesive as depicted in Fig. 10 instead of welding as in the case of UAE process.

In recent times, the LOM techniques have experienced several modifications and development that made the technique capable of manufacturing materials with complex phases. Using the LOM technique as its basis, a 3D printing process for laser-induced graphene (LIG) was developed by Luong et al. [57]. The developed process combined the LOM technique and the process of subtractive laser milling, in order to improve refinements, and the graphene foam is

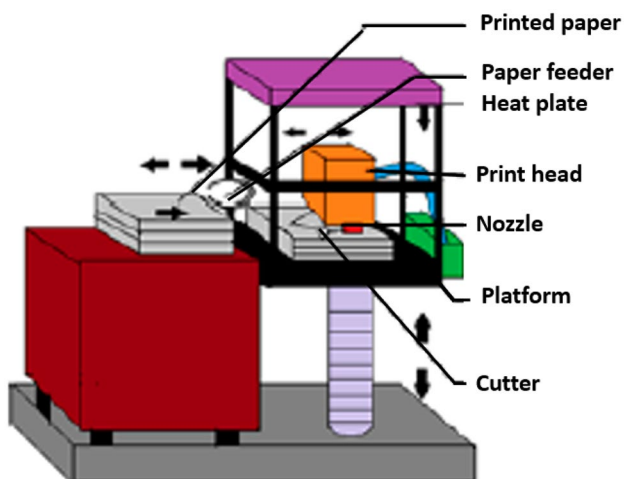
**Fig. 10** Lamination object manufacturing technique that requires adhesive for bonding



developed. The use of these combined processes or techniques helped in the fabrication of polymer composites with good mechanical strength, and electrical conductivity, that are suitable for use as energy storage, and/or can be used for electronic sensor applications. Thermal residual stress development and possible deformation resulting from layer mismatch and gradient cooling are the common issues associated with LOM fabricated products [58]. The feasibility of preparing silicon nitride ceramics components using aqueous tape casting and the LOM technique was effectively demonstrated by Liu et al. [59], and the produced components displayed a good flexural strength. In a related study, MAX-phase ( $M_{n+1}AX_n$ ) components were synthesised by Krinitcyn et al. [60] using the LOM process. A frozen-slurry-based LOM-slurry was developed by Zhang et al. [61]. The LOM-slurry was used in the fabrication of porous ceramic structures composed of powder alumina, water, and organic binder. Upon the crystallisation of the water in the LOM-slurry, the developed component is strengthened and can be cut with a laser in order to get the 2D pattern required before stacking them layer by layer, and then freeze-dried to give the needed porous structure.

Another AM process that requires each layer to be bonded with the previous one with the aid of adhesive as depicted in Fig. 11, and following the CAD data is the selective deposition modelling (SDM).

Based on the CAD data, the selective layer is cut and this procedure is repeated until the full part is completely fabricated. This method is unique in the fabrication of full-colour 3D parts, and it is also a good choice for concept models because the technique allows the production of highly stable prototypes and tactile models. This technique was used by Hung et al. [62] to prepare ordered flexible layers of graphene oxide on a modified polyacrylonitrile substrate.



**Fig. 11** AM process that requires bonding before selective deposition modelling

In the plastic sheet lamination (PLS) technique, rather than using an adhesive, heat and pressure are used since the process entails melting the sheets together. Parts produced using the sheet lamination process are not suitable for use in structural designs but are frequently used for visual models and aesthetics. Metals like copper, aluminium, titanium and stainless steel are used in the UAM technique. For the fabrication of composite parts using materials such as fibre, aramid fibre, and fibreglass in conjunction with metal fibre such as aluminium, titanium and steel, selective lamination composite object manufacturing (SLCOM) technique is preferably used, as this technique is used under low temperature and does not require much energy to bond the different materials. This technique is relatively cheap because it uses standard materials. Nevertheless, some of the disadvantages of this manufacturing technique (SLCOM) are [63–66]:

- Post-processing is required to obtain the desired product
- The resolution of the part is a function of the thickness of the sheet
- Profused waste is generated in the process
- Available material options are limited
- The strength of the bond formed is dependent on the used laminating technique

### 3 AM of polymers

Additive manufacturing of polymers can be done using different polymeric materials in different compositions and forms, composites, nanocomposites, discontinuous/continuous fibre-reinforced thermoplastics composites, and hybrids. A breakdown of some of the commonly used methods and processing techniques for 3D printing of polymer in the polymer industries and the major players in the 3D printing manufacturers are depicted in Table 1.

Polyamide (Nylon), polylactide or polylactic (PLA), acrylonitrile butadiene styrene (ABS), polycarbonate (PC), nylon 12, epoxy resin and glass-filled nylon are the most used polymer materials in FDM processes. Commercial polymers such as polycarbonate (PC) and ABS-M30 are known for their popular use in the FDM process [67]. Nevertheless, polyamides such as thermoplastic polyurethane (TUP), Nylon PA 120 and Nylon PA 11 are commonly used in multi-jet and SLS processes [68].

The advancement in AM has seen the development of different photo-active polymers that are used in the SLA technique. A typical example of such photo-active polymer developed, and used in the SLA technique, is the Somos<sup>®</sup> ProtoGens. It is an ABS-like liquid photopolymer SLA resin that first demonstrated different material properties under a machine-controlled exposure [1]. Studies have shown that the high-temperature resistant ABS-like photopolymer

**Table 1** Additive manufacturing techniques used for polymer composites fabrication, and the 3D-printer manufacturers and their locations

Product formation methodology	Additive manufacturing technique	Manufacturer of 3D-Printer and location	Polymer used
Matrix extrusion	Fused deposition modelling, FDM	Ultimaker B.V (Netherlands), StrtaSys (USA), Bigrep (Germany), Markforged (UK), Markerbot (USA), Intamysys (China), Robze (Italy), Tractus-3D (Netherland), Raise-3D (USA)	PC, PEEK, Nylon 12/carbon Fibre, PLA, ABS, PEI, HIPS, ASA, TPU
Vat polymerisation	Stereolithography, SLA	Origin (USA), Nexa 3D (USA), Photocentric (UK), Asiga (Australia), Envisiontec (USA), Prodways (France), Carbon (USA), Formlabs (USA), 3D-Systems (USA)	WaterClear Ultra 10,122; NeXt (white), Somos <sup>®</sup> ; EvoLve 128 (white), Somos <sup>®</sup> ; Element (clear), Somos <sup>®</sup> ; BioClear (clear), Somos <sup>®</sup> ; 9120 (off white), Somos <sup>®</sup> ; stereolithography (SLA) materials, Somos <sup>®</sup> ; liquid UV-curable photopolymers Somos <sup>®</sup>
Powder bed fusion	Multi-jet fusion Selective laser sintering, SLS	Multi jet Fusion Farsoon Technologies (China), Sinterik SA (Poland), Prodways (France), Formlabs (USA), 3D-Systems (USA), Eos, GbbH (Germany)	Elastic TPU, Polyamide (PA12/Nylon 12) Polyaryletherketones (PAEK), thermoplastic elastomers (TPE), polystyrenes(PS), polyamides(PA), acrylic styrene (PMMA/PS), nylons (polyamide (PA)), polycarbonate (PC) in their powdered form
Material jetting	Material jetting	Mimaki (Japan), 3D-systems (USA), Objet (USA)	Digital ABS, Durus, Tango, Vero

prefers the opportunity of accurately building 3D parts for different general and specific applications. It is worth noting that their processing attitude makes them suitable for automotive, aerospace, medical, and electronics markets, where highly durable and accurately modelled concepts whose parts are humidity and temperature resistant are required [1]. Another resin that is also available in commercial quantity for use as an optically clear stereolithography material is SOMOS water clear ultra 10122A. This resin is used for colourless functional parts that require excellent temperature resistance. Ultraviolet light-cured liquid photopolymer droplets are used to build parts in the polyjet techniques, and the polyjet materials available in commercial quantities (i.e. biocompatible plastics) can be used in medical technology. In addition, different materials can be blended/mixed together to give a component with new properties. Materials such as Vero (a smooth surface rigid plastic that comes in different colours and transparent Stratasys), Durus White (polypropylene material simulated from Stratasys), RGD 525, digital simulated ABS, and Tango (a soft rubber-like polyjet material) are often used in the polyjet printing process. Depicted in Table 2 are the typical properties of additive manufactured components that are developed using the different commercial polymers obtained from the EOS GmbH, USA, Stratasys, and SYS datasheets.

Rationally, the polymers used in additive manufacturing processes can be classified as:

- Thermoplastics
- Polymer matrix composites
- Particle-reinforced polymer composites
- Thermo-responsive polymers,

**Table 2** Properties of polymers (commercial) used in AM processes

Polymers	Material properties		
	Density (kg/m <sup>3</sup> )	Tensile modulus (MPa)	Tensile strength (MPa)
PLA	22	1627	1050
ABS	50	2347	1240
PC	40	1944	1200
Nylon 12	32	1282	950
PA 11	48	1600	990
PA 12	48	1650	930
Water Clear Ultra 10,122	56	2880	1130
Protogen O-XT 18,240	68	2960	1160
Durus White	30	1200	1170
RGD 525	70	3500	1180
Vero	65	3000	1190

- Fibre-reinforced polymer composite
- Thermoplastic elastomers composites and
- Nanocomposites

The advancement in the additive manufacturing of these polymers and their composites are presented below in subsequent sections.

### 3.1 Additive manufacturing of thermoplastic

As a plastic with the ability to soften upon heating and solidify upon cooling, thermoplastics are melt-processable plastics that retain their inherent properties during cooling and solidification [69]. Over the years, thermoplastic parts are produced using traditional processes such as injection moulding and extrusion. With the advent of 3D or AM technologies such as SLS and FDM, the creation of complex thermoplastic parts is now possible. Using this new manufacturing technique, the thermoplastic is heated until it attains a malleable state after which it is extruded onto a platform where it is allowed to solidify [70]. High stability, good tolerance at high temperature, high strength and rigidity are some of the reasons for the use of thermoplastics; and nylon, PLA, PE, ABS etc. are some of the commonly used thermoplastics [71]. The development of AM techniques such as FDM, SLS and SLA has made it possible for the development of components suitable for use in general-purpose, special biomedical and engineering applications. FDM technique was used by Gkartzou et al. [72] in the 3D printing of blends of bio-based polylactic acid (PLA), and the parameters such as the extrusion temperature, print speed and fibre width, which are the major contributor to the imposed stress on the melt, were examined. The use of the SLS technique in the powder densification and thermal modelling of amorphous polycarbonate was reported by Childs et al. [73]. In their report, three strategies, namely analytical strategy, fixed mesh finite element and adaptive mesh finite difference, were investigated, and the ability of the strategies to predict the physical process behaviour was evaluated by comparing the three strategies with experimental results. AM technique was used by Lee et al. to develop polypropylene fumarate (PPF) that possesses different porosity in order to investigate its suitability for use as scaffolds in tissue engineering [74]. In another study, polymethylmethacrylate (PMMA) microfluidic devices suitable for use in biomedical applications were developed by Matellan and Armando [75] using AM technique. AM technique has also been used successfully in the 3D printing of water-soluble scaffolds using PDMS microfluidic chamber [76]. The same technique has also been used successfully with polymer systems to produce craniofacial and dental scaffolds [77]. Through 3D printing of thermoplastic, Valtonen et al. replicated nasal cavities and determined their suitability for clinical applications by

studying the ease of air passage through them [78]. The use of polymer-based AM technique has proven to be one of the ways of fabricating complex biomedical structures for biomedical applications in a cost-effective way. Aside from biomedical applications, polymers are also reinforced with other materials to improve the mechanical properties, thus making them suitable for other applications.

### 3.2 Additive manufacturing of polymer matrix composites

Generally, polymers are preferred in additive manufacturing processes because of their unique adaptability to different processes and the flexibility they gave in the fabrication of customised products with complex geometries with a high degree of accuracy [79]. On their own, polymers possess inferior mechanical properties; as such, they are not suitable for load-bearing applications. In a bid to overcome this challenge and make polymers suitable for a number of load-bearing applications, research has been conducted on ways to improve the mechanical properties of polymers by developing polymer matrix composites (PMCs).

In general, a typical polymer matrix composite (PMC) is comprised of different fibres which can either be short or continuous, which are held together or bound together by an organic polymer matrix. The use of fibres and particles to reinforce polymers is a way devised to increase the mechanical properties of polymers such that the newly developed PMC is found useful in load-bearing applications. The polymers can be synthetic or biomaterial resins [80]. Carbon-based materials are often used as reinforcement in polymers and the FDM technique is mostly used in the fabrication of such composites. Other reinforcement materials commonly used in the FDM technique to reinforce polymers are carbon nanofibers, TiO<sub>2</sub>, graphene, montmorillonite, etc. For the SLA technique, the frequently used reinforcement includes BaTiO<sub>3</sub>, TiO<sub>2</sub>, and graphene oxide carbon nanotubes.

Several studies have been conducted by different researchers on the development of polymer matrix composites. In one of the researches conducted, Sánchez et al. [81] developed a carbon fibre-reinforced acrylonitrile styrene acrylate (ASA). On studying the mechanical properties of the developed PMC, a 350% increase in flexural Young's modulus and a 500% increase in the thermal conductivity were observed by the authors when 20 wt % carbon fibre (inclusion) was used to reinforce ASA.

Glass fibre is another commonly used reinforcement in PMC development because the glass fibre orientation and its composition with polymers make it ideal for use in load-bearing applications. It has been reported that “the functional composites of glass fibre-reinforced composites are equal to steel, and it has a higher stiffness than aluminium” [1, 82]. The influence and characteristics of

layer thickness and orientation of glass-reinforced polymer composite were studied by Carneiro et al. [83] using the FDM technique. They discovered that the FDM technique used is suitable for the fabrication of small components and parts, and desired interfacial adhesion was identified as one of the crucial requirements the reinforcement must meet in order to achieve an enhanced fibre-resin matrix affinity and also increase the durability of the developed PMC [84]. To achieve this, coupling agents are introduced to the fibres with the polymer. Polyethylene-graft-maleic anhydride reinforced with short sisal fibre composites was studied by Fernandes et al. [85]. The adhesion between the matrix and the fibre in the study was improved by treating the fibre with alkali before producing the composite with the aid of a twin-screw extruder and a compression mould. An improvement in the tensile and flexural properties of the developed composite was observed when 10 wt % sisal fibre and 2 wt % coupling agent were used. To improve composites production via additive manufacturing, four design principles, namely, position and fixation, layup and handling aids, structural handling aids, and post-processing aids, were suggested by Turk et al. [86]. Table 3 shows some of the materials and techniques used in the development of polymer composites and the properties of the composites that were enhanced due to the technique used.

### 3.3 Polymer composites with particle reinforcement

Polymer composites with particle reinforcements consist of particles of a particular material that is dispersed in the matrix of another material. The dispersed particles in the matrix vary in terms of shape, size, and morphology. In general, the shape of the particles is spherical, polyhedron, oval or irregular, and these particles are added to the liquid matrix during the process of forming the composite. The mixed particle and liquid matrix solidifies and is grown through procedures like pressing together of the composite, age hardening, and inter-diffusion via powder processing etc.

Processes like cryogenic ball milling, wet grinding-rounding, emulsion precipitation, dissolution-precipitation and spray drying are used during the preparation of the composite powder [91], and the size distribution, particle distribution, morphology and constituent within the matrix are determined by the process of preparation. Based on the mechanism of strengthening, particle-reinforced polymers are classified into particulate-reinforced and dispersion-strengthened composites [92]. In particulate-reinforced composites, there is a dispersion of coarse particles of the reinforcement in the matrix material, while in the dispersion strengthening composites, the size of the particles is usually small (in the range of 0.01–0.1 μm) and the strengthening of the composites occurs at the molecular or atomic level [1]. For the particulate-reinforced composites, interaction (interfacial) between

**Table 3** Additive manufacturing techniques used, and the corresponding properties enhanced in the composites

Technique used	Materials used	Properties enhanced	Reference
Multimaterial AM	Multifunctional materials that are suitable for mimicking the mechanisms of unibody robotics	The developed composite is MR-compatible, with excellent performance	[87]
Multimaterial stereolithography	Multi-chip modules that have enhanced on-package dielectric lens suitable for mm-wave applications	3D printing of multiple materials that possess different dielectric constants at different optical resolutions. These allow for the entire formation of new structures that can be integrated into a system-on-package solution, and used for mm-wave applications	[88]
Multi-nozzle AM system	A structure of hard and soft materials padded sandwich	An improvement in the printing performance as compared to the traditional FDM	[89]
Digital light synthesis	Suspended silica nanoparticles (functionalised) in poly(dimethylsiloxane) matrix	The use of resin and silica nanoparticles with a 0.15 mass fraction allows for printing at standard temperature and pressure (STP)	[90]
FDM	Polypropylene	The use of FDM for the fabrication of small series of components/parts that favourably compete with the conventional techniques. The FDM technique produced parts with improved mechanical properties, and with no restrictions in prototype production	[83]
Large format AM	Acrylonitrile styrene acrylate (ASA)	The carbon fibre composites displayed higher performance as compared to the raw ASA polymer (i.e. the composite with 20 wt % carbon fibre gave a 350% and 500% increase in the flexural Young's modulus and thermal conductivity respectively as compared with the pure ASA)	[81]

the matrix and dispersed phase is crucial in the determination of the properties of the developed composite.

Stereolithography technique was employed by Korhonen et al. [93] to fabricate graphene-based composite. To reduce the graphene oxide in the fabricated composite to graphene and improve the electric conductivity of the composite, the composite was pyrolyzed, and the achieved electric conductivity obtained was in the semiconductor range. Supramolecular polymer was 3D printed by Rupp et al. [94] and the effect of phase separation and nanoparticles on the printability was examined. The supramolecular polyisobutylene polymers displayed rubber-like behaviour but the polyisobutylene nanocomposites fabricated via the mixing of silica nanoparticles (5–15 wt %) of approximately 12 nm in size showed high structural stability and improved shape persistence.

In a bid to enhance the strength of particle-reinforced composites, a hybrid model was formulated by Abedini and Chen [95]. The model showed that the nonlinear behaviour a composite exhibits, when it is under uniaxial tension, strongly depends on the volume fraction and particle size. In a related study, Yuan et al. [96] used thermoplastic polyurethane to fabricate soft and flexible 3D metamaterials; and also used the SLS process to fabricate multi-walled carbon nanotubes (CNTs)-reinforced polymer composites. The developed composites displayed better heat absorption and heat conductivity; and the addition of CNTs was observed to enhance the toughness, tensile strength, and elongation of the composites [97]. Table 4 shows a summary of the additive manufacturing techniques used, and the general trend in the properties of particle-reinforced polymers.

### 3.4 Fibre-reinforced polymer composites

The building materials of fibre-reinforced composites (FRC) consist of three components, fibre, matrix, and the interface. Through the combination of two or multiple materials with different properties, composite materials are produced with improved properties that cannot be achieved by either the matrix or fibre alone. The strength, chemical stability, modulus of the fibre, and the quality of the interfacial bond that exists between the fibre and matrix determine the mechanical properties of the fibre-reinforced composites.

Stereolithography (SLA), laminated object manufacturing (LOM), fused deposition modelling (FDM), ultrasonic additive manufacturing (UAM), additive gypsum printing manufacturing, selective laser sintering (SLS), and fibre encapsulation additive manufacturing (FEAM) [97, 98] are the additive manufacturing techniques used for the fabrication of fibre-reinforced polymer (FRP) composites; and carbon fibres are the most commonly used reinforcement because they produce composites with low thermal expansion, low density, and improved thermal conductivity. The use of AM

**Table 4** Additive manufacturing techniques used in the fabrication of particle-reinforced polymer composites, and the corresponding properties of the composites enhanced

Technique used	Materials used	Properties enhanced	Reference
SLM	Auxetic foams made of thermoplastic polyurethane (TPU) with high porosity	The auxetic properties of the composites are retained over a wide range of deformations. Fast recovery when subjected to repeated compressive load	[96]
Thermally reversible and shear-induced dissociation of supramolecular polymer network	Supramolecular polymers (having linear and three-arm star), with hydrogen bonds attached and their nanocomposites	The fabricated supramolecular PIB polymers display a rubber-like behaviour, and are capable of forming self-supported objects that can be 3D printed at room temperature and below, thus allowing the strand diameters to the polymer up to 200–300 µm	[94]
Stereolithography	Graphene oxide/polymer composites	3D structure was successfully designed using this method, although, further optimisation is required in case of practical applications due to the brittleness and the high shrinkage associated with the pyrolysed 3D composite fabricated	[93]

techniques in the fabrication of these composites helps in the significant reduction in the production time as compared to the conventional methods. The use of this technique allows for a large build envelope and reduction of warping [99].

A capillary-driven AM technique for the fabrication of continuous carbon fibre composites was reported by Shi et al. [100]. The dynamic approach makes it possible to control the degree of curing and viscosity of the composites, and allows simultaneous infusion and curing of the composites, thus resulting in the in situ solidification of the developed composites. A high fibre fraction (58.6%), 95% degree of curing, high mechanical strength with an approximate value of 810 MPa and modulus of elasticity of 108 GPa were obtained when this method was used in printing the composite. Despite the wonderful attributes such as low cost and design flexibility associated with the use of this AM technique in the fabrication of composites, they are limited by low stiffness and strength, when compared with conventional methods of manufacturing composites. In an attempt to address these limitations, a “vibration integrated auger extrusion system” was developed and used in the fabrication of carbon fibre-reinforced composites [101]. This AM technique allows the fabrication of short fibre-reinforced composites and also makes it possible to manufacture composites with improved flexural strength, compression strength, stiffness and higher volume ratio of fibre. Another frequently used fibre in the fabrication of FRC aside from carbon and glass fibre is Kevlar [102]. FDM technique was used by Dickson et al. to develop fibre-reinforced composites and also study the effect of using continuous carbon fibre, glass fibre and Kevlar [103]. The influence of fibre type, fibre orientation and volume fraction of the fibre in the composites was studied, and it was discovered that the composites reinforced with glass fibres gave the highest tensile strength. Table 5 shows the AM techniques and the properties of the developed composites when a polymer is reinforced with fibre.

### 3.5 Nanocomposites

The ease of combining nanotechnology and additive manufacturing has made it possible for the fabrication of 3D parts with multiple functionalities and optimised properties. Through the incorporation of nanomaterials such as carbon nanotubes, there is a significant improvement in the mechanical strength, electrical conductivity, chemical, and electro-mechanical sensitivity of the developed nanocomposites [105]. Although a number of 3D nanocomposites are fabricated using the available AM technologies, the fabricated nanocomposites are used in different fields of applications such as micro-electronics, microelectromechanical systems (MEMS), tissue engineering, engineered materials and composites, microfluidics, biosystems, and lab-on-a-chip, the common AM methods or techniques for the manufacturing/fabrication of nanocomposites include extrusion-based technologies, powder bed technologies, micro-stereolithography, and inkjet printing.

Stereolithography technique was used by Bustillos et al. [106] in the development of polymer and boron nitride nanoplatelet composites, and the influence of nanoplatelets on the nanocomposites was evaluated. During the evaluation, it was observed that the damping, compressive strength and micro-hardness of the nanocomposite increased with the addition of the nanoplatelets. It was further discovered that during curing, the interaction of the nanoparticles with the laser wavelength is the most crucial factor to consider when using AM technique to manufacture a composite with improved functional properties.

A 3D printing technique was employed by Saleh Alghamdi et al. [1] to develop nanocomposites consisting of an ultraviolet-curable polymeric resin reinforced with different inorganic fillers. The effect of the concentration of the filler on the rheological properties of the nanocomposite was investigated, and the parameters that influence

**Table 5** Additive manufacturing techniques used in the fabrication of fibre-reinforced polymer composites, and the corresponding properties of the composites enhanced

Technique used	Materials used	Properties enhanced	Reference
Direct write additive manufacturing	Short fibre-reinforced thermoset composites	High flexural stiffness (about 53 GP), high flexural strength (about 401 MPa), high compression strength (673 MPa), and high fibre volume ratio (about 46%)	[104]
Dynamic capillary-driven additive manufacturing method	Carbon fibre composites	High mechanical strength of about 810 MPa and 108 GPa modulus of elasticity, high fibre volume fraction (about 58.6%) and a high degree of curing (about 95%)	[97]
FDM	Glass fibre, Kevlar, and Continuous carbon-reinforced composites	As fibre content in the glass specimen approached 22.5%, maximum tensile strength was observed. Further increase in the fibre content up to 33% only resulted in a slight increase in the strength of the composite	[99]

optimal printability were determined. A nanocomposite (polydimethylsiloxane (PDMS)) with strain sensing and electrical conductive properties was developed by Abshirini et al. [107]. In the nanocomposite, a multi-walled carbon nanotube was distributed uniformly in the PDMS and a scanning electron microscope (SEM) was used to evaluate the microstructural features of the nanocomposite. By subjecting a specimen of the developed nanocomposite to cyclic tensile loading, the strain sensing ability was evaluated, and the developed nanocomposite was observed to have high strain fidelity. In other related studies, a photothermally responsive hydrogel graphene oxide-reinforced poly(N-isopropyl acrylamide) was 3D printed by Zhang et al. [108]. Using stereolithography technique, nanocrystal-reinforced methacrylic cellulose was 3D printed by Wang et al. [109]. The developed nanocomposites were observed to have good particle dispersion, better mechanical properties and thermal stability. With the aid of high conductive CNTs and PLA, Chizari et al. [110] fabricated nanocomposite scaffold structures for liquid sensor application.

## 4 Applications of additively manufactured or 3D printed polymer composites

The usage of additively manufactured polymer composites spans across several industries such as aerospace, electronics, biomedical, construction, and textile industries. Worthy of mentioning is the use of this composite to fabricate flexible components/parts such as textile materials and wearable electronics components. Several studies have been conducted in different fields of endeavour on the applications of additively manufactured polymer composites. Some of the research conducted on the application of these composites in different field are discussed in subsequent sections.

### 4.1 Biomedical applications

The biomedical fields have benefited tremendously from the use of AM to manufacture polymer composites suitable for different biomedical applications, ranging from the manufacturing of customised biomedical tools and medicines to the manufacturing of different organs. The biomedical applications of polymeric materials are categorised into soft and hard polymers, and these polymers are further categorised into biodegradable and non-biodegradable polymers. In biomedical applications (i.e. the human body) where tissues are expected to grow inside the body, and the biomedical parts inserted inside the body are no longer required, soft biodegradable polymers are used. After a while, this biodegradable polymer degrades in the human body.

On the other hand, hard non-biodegradable polymers are employed in the biomedical field as structural implants, and

the parts they are used to create are expected to remain in the body for as long as possible. Polylactic acid (PLA), polyglycolic acid (PGA), polycaprolactone (PCL), and polydioxanone (PDO) are some of the hard-synthetic biodegradable polymers frequently used in biomedical applications [111], while the polymers of polyaryletherketones (PAEKs) are parts of frequently used non-biodegradable hard polymers. Dental implants, prosthetics and surgical tools are some of the applications of hard polymers in the biomedical field. Hydrogels such as poloxamer pluronic F127, with unique thermal properties, polyethylene glycol (PEG), elastomers and acrylate-based hydrogels are parts of the soft polymers frequently used in biomedical. The biomedical applications of these soft polymers include drug delivery, tissue engineering and bioprinting, soft surgical models, and tissue phantoms [71].

Scaffolds suitable for cardiac tissue engineering were additively manufactured by Ho et al. [112] through the reinforcement of polycaprolactone with CNTs. An AM technique was developed by Ramírez et al. [113] to fabricate composite materials with improved mechanical properties using FDM/FFF methods. The use of AM technique to fabricate components for dental applications such as prostheses, maxilla-facial, and orthodontic appliances was studied by Jockusch and Özcan [114].

### 4.2 Applications of AM in the electronics industry

The electronic industry has benefited immensely from the additive manufacturing of PMC, especially in embedded electronics. Two methods are used in the fabrication of PMC in embedded electronics. The first method entails the prefabrication of discrete components that are transferred directly into the substrate using a laser, while the second method involves the laser printing of the functional components such as interconnects, passives and actives of the substrate [115]. The use of carbon nanotube-based materials for 3D printing of functional electronics parts was studied by Goh et al. [116]. Using AM technique, flexible supercapacitors were manufactured from a polypyrrole-MnO<sub>2</sub>-carbon fibre hybrid by Tao et al. [117], and the active materials of the composite displayed high energy management potential.

Silicon rubber and carbon black nanocomposite were used by Wang et al. [118] to develop a thin flexible pressure sensor. In a related study, a flexible and stretchable high-strain sensor that consists of graphite flakes or CNTs, which are distributed randomly in a natural rubber substrate, was reported by Tadakaluru et al. [119]. According to their finding, the developed CNT sensor gave 620% strain, while the graphite sensor gave 246% strain. The values obtained for the CNT and graphite sensors are approximately 120 and 50 times, respectively, greater than the conventional metallic strain sensors. The feasibility of manufacturing flexible



3D printed antennas from conductive acrylonitrile butadiene styrene (ABS) using additive manufacturing techniques was investigated by Mirzaee et al. [120]. The outcome of the study showed that conductive ABS can be suitably used for the 3D printing of electromagnetic structures.

### 4.3 Aerospace applications

In the aerospace and defence industries, laser-based additive manufacturing or 3D printing techniques are widely used because of their exceptional ability to fabricate complex geometries that are cost-effective. FDM, SLA, and jet-type processes such as polyjet are the most used polymer-based AM processes in aerospace applications. Research conducted on parts fabricated using SLA and FDM additive manufacturing techniques showed that the SLA-fabricated products are stronger than those fabricated using FDM, thus making SLA-fabricated products more suitable in the aerospace industries, for casting purposes [121]. Customised parts made with high strength and low weight materials are required in the aerospace industries and the parts can be easily manufactured on demand using additive manufacturing techniques. The Desert RATS (a Rover) designed by NASA have about 70 of its parts manufactured via AM. Some of the additively manufactured Desert RATS parts are the front bumpers, flame-radiant housings and vents, complex electronics, camera mounts, large pod doors, etc., and the parts are made of PC, PCABS and ABS, produced via the FDM technique [122]. The AM application in unmanned vehicles was studied by Goh et al. [123] and the ability to print multiple and smart materials, multifunctional structures printing, on-demand printing, on-site printing etc. are some of the potentials of AM in unmanned vehicles. The use of polymer nanocomposites (PNCs) in the aerospace industry has attracted a lot of attention, particularly in the fabrication of composites, suitable for aerospace applications. The composites are made of differently shaped nanofillers such as fibres and platelets, in the polymer matrix [124]. As compared to conventional composites, polymer nanocomposites give improved properties such as higher thermal performance, modulus, atomic oxygen resistance, resistance to molecular penetration, and improved ablative performance [125]. Clay, graphene oxide, graphene, and carbon nanotube are the nanocomposites that are commonly used for aerospace applications [126]. Because these nanocomposites are unable to attain the necessary strength requirements for use in aerospace structures, hybrid composites also known as multiscale composites are developed by combining conventional composites and nanocomposites in order to attain the required strength needed for aerospace applications [127]. In a bid to address the effect of damage associated with composites materials developed for aerospace applications, self-healing carbon fibre-reinforced polymer composites were

developed by William et al. [128]. Their study showed that there is self-healing of the composites, as observed through the flexural strength recovery when the laminate filled with resin at 70 and 200  $\mu\text{m}$  hollow glass fibres (HGF) spacing.

### 4.4 Applications in textile industries

The ease of improving the complexity and functionality of polymer composites using additive manufacturing has encouraged its use in the textile industry. AM also allows for the designing and development of novel polymer composite materials suitable for textile applications [129]. In the manufacturing of textiles via additive manufacturing techniques, the main focus must be on producing important textile properties such as strength, softness, porosity and flexibility [130]. Using AM techniques for textiles makes it possible to produce customised clothing, and also to carry out well-coordinated tasks during the production of components that requires transformation from solids to textiles, and incorporate additional functionality through design as obtained in additively manufactured optimised footwear [131]. 3D printing on textile substrates was investigated by Korger et al. [132] and they discovered that adhesion and good stability are crucial criteria to consider when 3D printing on textile substrates after conducting abrasion resistance and separation force test on the prints. Using the FDM additive manufacturing technique and different thermoplastics, textile-based structures were manufactured and investigated Melnikova et al. [133]. According to the authors, great adhesion properties can be obtained if the textile is thick or if the surface of the textile is hairy or rough, and this good adhesion result obtained in their study is attributed to the form-locking connections of the printed polymer to the fibres inside the textile structure, as well as on top of the textile, as this allows sufficient opening, which in return aids the penetration of molten polymer.

The adhesion of polymers such as nylon, ABS, and PLA on different types of fabrics was investigated by Pei et al. [134], and PLA was observed to have the best adhesion properties. Stab-resistance characteristics of planar samples manufactured with 100% virgin nylon (PA2200) or 50:50% virgin and recycled nylon were explored by Johnson et al. [135]. The obtained results were then used to additively develop an articulated laser-sintered scale textile that gives a maximum knife penetration that is within the acceptable limit. Leist et al. [136] explore the possibility of additively manufacturing textiles from shape memory polymer. To achieve this, PLA was combined with nylon fabric to produce smart textiles that can be trained thermomechanically to produce different shapes, and also return to their initial shapes upon exposure to high temperatures.

Maiti et al. [137] conducted a study on the use of conductive polymers like thiophene, pyrrole, aniline and their

other derivatives to develop flexible and electro-conductive non-metallic textiles.

## 5 4D printing

As an extension of 3D printing, 4D printing makes it possible for the properties and shapes of printed objects or articles to be changed temporarily. The 4D printing technology involves the incorporation of 3D with smart materials in a bid to develop components that are capable of assuming multiple configurations upon the application of environmental stimuli such as mechanical stress, magnetic field electric field, chemical agent, pH and temperature [138]. With its first highlight in 2013, the technology enables dynamic structural development of complex items with the ability to undergo shape transformation. Smart components capable of self-adapting, self-assembly and self-repairing are some of the potential qualities of components fabricated using 4D printing [139].

Shape memory polymers and hydrogels are some of the commonly used materials for 4D printing [140]. Being a new technology, 4D printing is faced with many challenges that need to be overcome if it must attain its full potential in the technological world. Some of the challenges faced by this technology include the reduction in the strength (mechanical) of the materials developed and a significant delay in response to stimuli, which eventually lead to slow shape change rates [141].

In order to 4D print a part, four major approaches are used, and they are [142]:

- **Self-assembly of elements:** Elements self-assembly makes it possible for automatic self-assembly of a 3D printed component upon the application of certain external conditions. The use of this method is common in biomedical applications (i.e. the method allows for the supply of active components via a tiny hole inside the human body, and the active components are assembled inside the body during surgery) [143].
- **Bi-stability:** Structures with two or multiple stable positions under certain conditions also possess zero degrees of freedom and are used in the bi-stability approach. While the structure is properly induced for slight deformation, it has the ability to change from one stable position to another [144].
- **Deformation mismatch:** In the deformation mismatch 4D printing approach, the deformation mismatch is induced using physical properties such as thermal expansion coefficient and swelling ratio.
- **Shape memory effect:** After deformation, shape memory effect (SME) takes place in some of the materials as

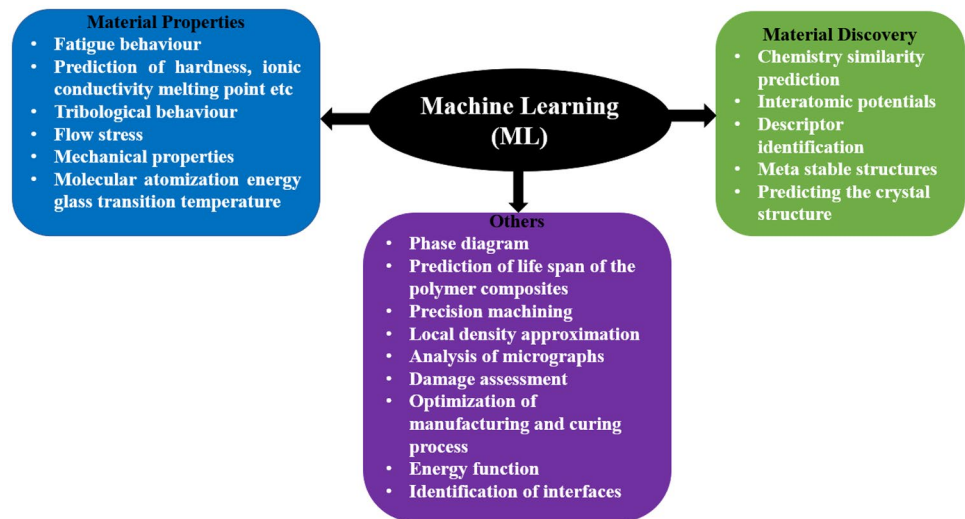
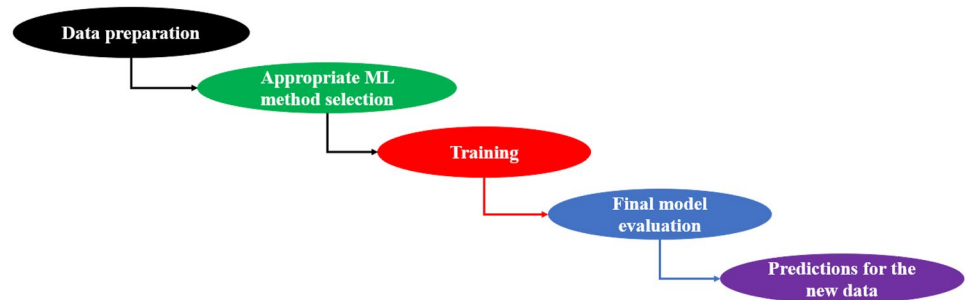
they are able to return to their original shapes upon the application of the appropriate stimuli [145]. The magnitude of shape deformation or change in some materials is dependent on the proportion of the applied stimuli known as the change effect (SCE) [146].

With the emergence of 4D printing, there are boundless opportunities in terms of the creation of smart objects in different areas like the medical and biomedical fields, robotics, manufacturing industries and aerospace. [147–155]. 4D printing has shown the capability to effectively fabricate smart material tissue interfaces for use in biomimetic and bioinspired devices [153]. A major challenge facing the use of 4D printing in biomedical applications is the limited availability of cost-effective, smart materials suitable for printing biocompatible components. Hence, to sustain this AM technology, it is imperative to develop smart, multiple stimuli responsiveness and cost-effective biocompatible materials [154, 156].

The use of 4D printing for different applications has been studied by some researchers, one of which is a smart product of wearable textile developed by Lee et al. through 4D printing of shape memory polymer and nanocomposites [157]. 4D printing has also been employed in the field of electronics to fabricate electronics devices suitable for applications such as human/machine interference, soft robotics, and wearable electronics [158]. 4D printing has also been employed in the biomedical field to develop organs and tissues using biomaterials, biological molecules and cells [159]. Using 4D printing, stents that can be implanted in air passages to aid breathing in children were developed by Morrison et al. [160]; and customised porous radio-opaque shape memory polyurethane suitable for endovascular embolisation application was 4D printed by Kashyap et al. [161].

## 6 Machine learning application in additive manufacturing of polymer composites

As an artificial intelligence (AI) technique, machine learning (ML) allows for automatic learning of patterns between inputs and outputs [162]. These patterns learnt are based on the data used for training. Some of the ML algorithms such as artificial neural network (ANN) [163], linear regression [163], tree learners [162], and support vector machine (SVM) [164] can be used to identify the complex relationship that exists among nonlinear variables without specific knowledge or any physical models [165–167]. In the last two decades, there has been a gradual shift in the techniques used for the discovery and design of new materials. This shift is from purely computational techniques to methods that increase results reliability via the use of computational predictions that are validated experimentally [163]. Many

**Fig. 12** Machine learning application in polymer composites**Fig. 13** Machine learning algorithm for basic workflow

researchers, particularly in the area of polymer composites development have relied on the ML approach to decipher the appropriate process parameters that would give the optimal design [163, 168, 169]. Also, ML provides a wider scope for the efficient investigation of the behaviour of polymer composites with minimal experimentation or the development of computationally intensive and expensive models. The achievement of different tunable multifunctional polymer composites properties is possible through the use of ML, as this technique allows for different arrangement of the constituent components in different proportions.

In the field of polymer composites, machine learning has been identified as a powerful tool that can be used as an efficient predictive model that could give unprecedented insights into the exploration of the properties of the developed polymer composites beyond those obtained via the traditional experimental or computational analyses. Depicted in Fig. 12 is the illustration of the various applications of ML in polymer composites. This includes material development, material properties prediction and other areas such as prediction of the life span of polymer composites, and damage assessment.

The basic workflow of machine learning algorithms that fall under supervised learning follows the five steps approach described in Fig. 13. It is worth noting that any ML algorithm can be used depending on its suitability and efficiency for the particular problem to be solved.

## 7 Conclusions and future prospects

The recent advancements and achievements in the use of different additive manufacturing processes for polymers and their composites are reviewed in this article. From the summarised AM processes and some of the optimisation tools highlighted, there is no doubt that the future of AM of polymers and their composites in 3D printing from prototyping to the process of manufacturing robust components is promising. The continuous development of novel AM tools and techniques has paved the innovative ways for faster and smarter printing of bigger, weirder and complex polymer/composite structures with superior materials properties. Such superior properties include adhesion, flexural strength and modulus. These properties are identified as some of the

parameters that control the mechanical characteristics of additively manufactured polymer composites. The application of additive manufacturing of polymer composites in different fields such as electronics, construction, aerospace, biomedical, and textile industries is presented in this article and the additively manufactured polymer composites contributed tremendously to the different fields through the superior properties the developed composites displayed.

So far, AM has displayed high versatility in terms of manufacturing methodology which allow for the easy manufacturing of components with complex shapes, and the potential to transform traditional manufacturing completely in the future. The advances in AM are expanding daily, thus opening a new world of research, design and manufacturing, such that AM technology that was once considered only suitable for fabricating small and low-quality prototypes is now used in groundbreaking innovations [170–174] because the technology now offers engineers and designers the rare opportunity to push boundaries and develop composites with improved properties. Also, the use of AM technology has made it possible to fabricate bioinspired composites and electrodes that possess contradicting properties (a very porous structure with high flexural strength).

In addition, advancement in AM has offered engineers and designers unprecedented opportunities to fabricate complex structures with optimised performance enveloped that cannot be achieved using conventional methods. Hence, the advent of AM technology has redefined the way industrial designers and engineers think, as they can now reimagine new ways of creating components and products. With current progress made in additive manufacturing, the advent of machine learning, and the opportunities that the use of 4D printing tends to offer, there is no doubt the expansions and evolution of AM technology will definitely bring about new developments, reshape global supply chains, strengthen local networks, and reduce production losses drastically [175, 176].

Despite the numerous advantages the use of AM offers in the fabrication of polymer composites, the technology is limited by the availability of smart materials suitable for printing biocompatible polymer composites [150–153, 177]. Hence, there is still a dire need to develop more biocompatible smart materials with multiple stimuli responsiveness for biomedical applications [71, 154, 156]. Furthermore, much research attention still needs to focus on the other potential of using additive manufacturing techniques in design development and product manufacturing as this will help in sustaining the manufacturing method. The development of novel materials for thermoplastic elastomers can be achieved using blends of dimensional stable elastic polymers and ionomers such as sulfonates, phosphates and carboxylates. New materials with exceptional improved texture, flexibility,

strength and other relevant properties need to be developed for stimuli-responsive polymers.

Although the additive manufacturing process appears to be simple, it is a powerful process that offers endless possibilities in terms of product development, manufacturing, and applications. Despite being faced with a number of challenges that require urgent attention from the research world, the progress made thus far with AM, and the brighter future it tends to offer, is worth celebrating.

**Acknowledgements** The authors acknowledge Global Excellence and Stature (GES 4.0) and the Centre for Nanomechanics and Tribocorrosion, University of Johannesburg, South Africa, for their immense support.

**Author contribution** Smith Salifu: responsible for drafting, conceptualisation and writing. Olugbenga Ogunbiyi: responsible for review and editing. Peter Apata Olubambi: responsible for review and editing.

**Data availability** Not applicable.

## Declarations

**Ethics approval** Not applicable.

**Consent to participate** Not applicable.

**Consent for publication** All authors have read and agreed to publish the manuscript.

**Competing interests** The authors declare no competing interests.

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