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ADDITIVE TECHNOLOGIES OF POLYMER MATERIALS (Review)

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The paper gives the analysis of the state of the art of 3D technologies of polymer materials, which is based on publications presented both in open sources as well as in a wide range of scientific and technical journals, including the own experience of the authors in 3D printing using thermoplastic and thermosetting polymers. The history of additive technologies, state of the art and trends of the development of the market of three-dimensional printing are considered. The classification of the most widespread in the world technologies of additive manufacturing of products from polymer materials depending on the methods of processing plastics is offered, their short description is given, their features, advantages and disadvantages are presented. 27 Ref., 1 Table, 6 Figures.

Keywords: additive technologies, 3D printing, polymer materials

The current pace of industrial development requires the choice of production technologies that can be implemented in the shortest possible terms with the involvement of minimal monetary investments to obtain high-quality products as a result. Nowadays these requirements are met by additive technologies (Additive Manufacturing, Additive Fabrication or AM-, AF-technologies), which find an increased application in all spheres of human life and are one of the most interesting and promising areas of industrial production [1, 2]. The additive technologies are also called the technologies of layer-by-layer synthesis, 3D printing, 3D technologies, rapid prototyping, layer-by-layer modeling, digital production, etc.

The generalized term «additive» is accepted to denote a group of technologies that allow layer-by-layer creation of three-dimensional objects based on the data of CAD (Computer-Aided Design) model by gradual adding a source material to the future product, which differs from traditional methods of a part forming when an excess material is removed from the workpiece array applying mechanical treatment. The CAD model is a digital mock-up of the object, which should be formed with the use of additive technologies in the 3D printer. The three-dimensional CAD model can be both developed according to the own design applying the method of computer designing, as well as to create from the data collected by means of 3D-scanner.

The essence of Additive Manufacturing (AM) can be illustrated as follows.

The key feature of 3D printing is a significant reduction in the duration of a process chain from the idea or drawing to the process of manufacturing a product, and at the same time a reduction in labor, material and power consumption of manufacturing. Thus, additive manufacturing makes it possible to create final functional products directly from the designer or engineer through the computer and the printer without the use of additional technological operations.

Although 3D printing is considered to be one of the major discoveries of the XXI century, in fact, additive technologies appeared in 1984, when the American researcher Charles W. Hull, founder of the Company 3D Systems, developed the technology of layer-by-layer «growing» of physical three-dimensional objects from a photopolymer composition [3]. The technology was called «stereolithography» (STL). The author received a patent for his invention only in 1986, and a year later, the engineer presented to the public his industrial device for three-dimensional printing [4]. As far as the term «3D printer» appeared only in 1993, the machine of Charles Hull was called a «stereolithography apparatus.» Around the same time, other 3D printing technologies began to be developed. In 1985, Michael Feygin proposed the technology of lamination called LOM (Laminated Object Manufacturing), which allows binding individual layers of material using a special bead heated to a certain temperature [5]. In 1986, Carl Decard, an associate of the University of Texas, applied for a patent describing the process of rapid prototyping of products using the technology of SLS (Selective Laser Sintering) [6], the essence of which consists in layer-by-layer sintering of the powder material by a laser beam. In 1989, Scott Crump, later the founder of Stratasys, announced the development of fused deposition modeling (FDM) technology. When using this technology, the formation

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Basic methods and technologies of additive manufacturing from polymer materials

Method of forming 3D products	3D printing technology	Essence of technology or the principle of creating 3D objects
Extrusion	Fusing deposition modeling (FDM) or fused filament fabrication (FFF)	Successive deposition of layers of molten thermoplastic polymer material, which reproduce the contours of the digital model
Photopolymerization	Stereolithography (SLA or SL)	Use of photopolymer resins solidifying under the action of ultraviolet radiation
	Digital light processing (DLP)	Use of photopolymer resins solidifying when illuminated by a digital LED projector (DLP)
	Polyjet	Layered solidification of liquid photopolymer material under the action of ultraviolet radiation
	Multi jet modeling (MJM)	
Granulation	Selective laser sintering (SLS)	Successive sintering of layers of powder material using high power lasers
	Selective heat sintering (SHS)	Melting of thermoplastic powder layers by means of a thermal radiator
	Color jet printing (CJP) (formerly known as three-dimensional printing — 3DP)	Layer-by-layer gluing and painting of composite powder bases on plastics
Lamination	Manufacturing of objects by the method of LOM — laminated object manufacturing or PSL — plastic sheet lamination	Layer-by-layer gluing of film materials with the subsequent formation (cutting) of model by means of a laser beam or the cutting tool

of the object is carried out in layer-by-layer depositing, in other words by surfacing of a molten thread of thermoplastic material, which is extruded through the extrusion head onto the cooled platform. Then the process of development of the new technologies and improvement of the existing ones was accelerated and in 1993 a breakpoint came, which made additive methods of manufacturing products to be widely available, and the term «3D printing» to be known around the world. The students Jim Bradt and Tim Anderson of the Massachusetts Institute of Technology, who were operating on a dissertation on powder-based inkjet printing, introduced layer-by-layer synthesis into a conventional desktop printer [8]. In 1995, Tim and Jim founded the Company Z Corporation, which launched the technology at the market. In 2012, the technology was acquired by the Company together with the 3D systems Corporation. Today, under its brand the former models of Z Corporation are sold worldwide.

Nowadays, the market of three-dimensional printing is growing rapidly and replenished with new models of unique production equipment, the possibilities of which are almost limitless. Printed parts and assemblies of aircraft, printed car body, printed residential construction, printed clothing, printed medical implants represent far not the whole list of achievements of modern additive technologies. However, the most interesting thing is that it is possible to print using almost any materials: polymers, engineering plastics, composite powders, different types of metals, ceramics, sand, concrete, wood, and recently even food and biological substances [9–11]. Due to the availability and practicality 3D printing with the use of different types of plastic is the most common today.

All the technologies of 3D printing by plastic (Table) are based on four basic methods of processing polymer materials used in the industry for manufacturing of plastic products: extrusion, photopolymerization, granulation and lamination.

The most widespread 3D printing technologies which are used in everyday life (in the office) and industry are the formation of products applying layer-by-layer surfacing (FDM) and stereolithography (SLA). But, nevertheless, each of the abovementioned technologies finds its consumer, and also has advantages and features that arouse interest. Let us review each 3D printing technology in more detail.

FDM/FFF 3D printing. FDM layer surfacing is the most widespread 3D printing technology in the world, based on millions of 3D printers from the cheapest to industrial 3D printing systems. To create products applying FDM method of 3D printing, filament is used made of various thermoplastic materials, which are delivered in the form of coils. The filament can be of two standard diameters: 1.75 and 3 mm depending on the printer specification [12].

As in all 3D printing technologies, the first step in making a physical object is construction of a digital 3D model. The 3D model in STL format is transmitted to the 3D printer software. The program automatically (or manually by operator) places the model in the virtual space of the operating chamber. Then the program, if necessary, calculates the elements of auxiliary structures — supporting structures for overhanging elements of the object and calculates the required amount of consumables, as well as the time for «growing» the prototype. Before starting the process of printing, the model is automatically divided into horizontal layers and the ways of moving extruder (print head) are cal-

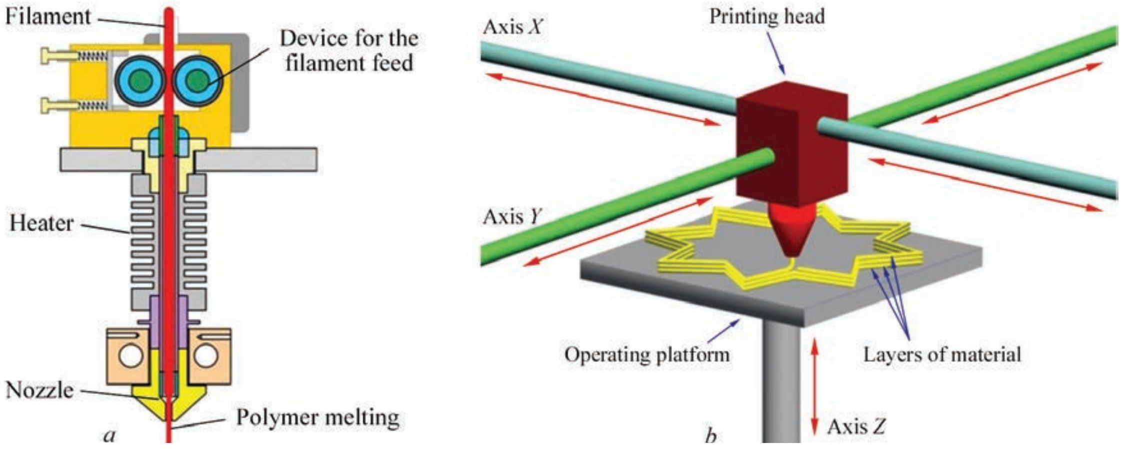

Figure 1. Scheme of extruder of FDM 3D printer (*a*) and the process of creating a three-dimensional model (*b*) by it [13]

culated. Extruder represents a device equipped with a mechanical drive for supplying filament, a heating element for its melting and a nozzle through which a direct extrusion-ejection of a molten polymer material on the surface of a product is carried out (Figure 1, *a*).

The received settings are saved, the model is converted into the control code for the 3D printer. Then the filament from the coil is unwound for its introduction into the extruder and directly the process of 3D printing is started: the extruder melts the filament and with a high accuracy in thin layers supplies the melt of the polymer material onto the operating surface of the 3D printer according to the printing algorithm and CAD of 3D-model. After deposition the layer, the polymer material is cooled and solidified, and the platform on which the object is formed is lowered to a level equal to the thickness of the deposited layer. The movement of the head and the platform in 3 planes (Figure 1, *b*) is set by an algorithm developed in advance with the help of a special software.

After the process of product manufacturing is completed, the auxiliary structures are removed (manually or dissolved in a special solution), and the finished product can be used in a printed form or subjected to any method of further processing.

The main advantages of FDM 3D prinitng are ease of using and the absence of special requirements for the room (suitability for using in the office); low cost of FDM printers and consumables; the possibility of using a wide range of types of plastic, depending on the needs and colors.

The main disadvantages of the technology are the need in creating supporting structures for overhanging elements, which after completion of printing have to be removed; low separating capacity, which leads to layering of the surface of the manufactured model; presence of thermal shrinkage is possible, which leads to a change in the size of a product after cooling.

SLA 3D printing. Stereolithography is not only one of the first 3D technologies of additive manufacturing in the world, but also one of the most pretentious [14–16]. That is why it is most in demand in the medical field, for example, for manufacture of implants, etc. As a consumable it uses liquid photopolymer resin. Under the influence of laser radiation on those parts of the photopolymer that correspond to the walls of the specified object, their layer-by-layer solidification and the formation of the finished product occur.

SLA 3D printing as well as printing by the FDM method, in parallel with construction of the object demands using of the supporting structures, which serve for fixation of a part of a product to the base of the platform and prevent deformation of the manufactured model in the presence of overhanging elements in it. There are two basic structures of stereolithographic 3D printers: conventional, which is used more often in industrial devices, and the so-called «3D SLA upside down printing», which is less common and found mainly in desktop 3D printers.

In the conventional variant of the SLA 3D printer the laser is located above, and the operating platform gradually lowers down (Figure 2). In a tank filled with a special liquid (mixture of photopolymer and reagent-solidifier), which looks like epoxy resin, a mesh platform is immersed to a depth of not more than 0.05–0.13 mm (this is precisely the thickness of one layer). After that, the laser is activated, as a result of which the liquid solidifies on certain parts of the material and sticks to the platform. Thus, the 3D printer creates the first layer of the product. Each layer represents a 2D image that is traced by a laser, according to the data embedded in a three-dimensional digital model. Then, the platform is lowered by one step, equal to the thickness of one layer, down (in the Z coordinate) and after leveling the surface of the liquid material, the laser is reactivated, continuing to build the next layer on the $X-Y$ coordinate axes. The cycles of forming layers are repeated before construction of a product.

The principle of 3D printing process used in desktop 3D printers is identical, with the only difference

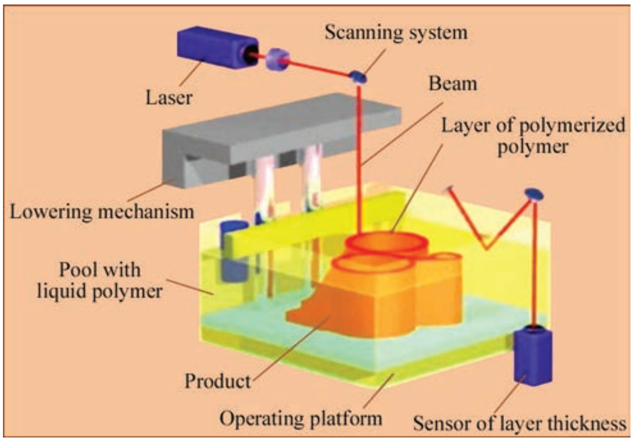

Figure 2. Scheme of SLA 3D printing process [17]

that the laser is located under the container with a photopolymer, and during construction of products, the platform does not lower, but gradually rises.

The feature, which is common to both variants, is that a product should be washed in special solutions to complete 3D printing, as well as irradiated with ultraviolet light. The first is required for the final cleaning of the manufactured model from photopolymer remnants, and the second for complete solidification of a product.

The main advantages of SLA 3D printing are high accuracy of construction of the finished product; the obtained model has a high compressive strength and ability to withstand temperatures of up to 100 °C; ability of producing complex models with preservation of small elements of decor; small amount of wastes; wide selection of consumables.

The main disadvantages of the technology are the massiveness of the equipment (large size and weight); high cost of the equipment and consumables; lack of possibility to use different materials in one cycle; shrinkage of the material during solidification, resulting in low flexural and impact strength; the need in ultraviolet treatment of the product after printing.

DLP 3D printing. Based on the SLA 3D printing method, several other methods of additive technologies were developed, one of which is DLP 3D printing. The difference between these methods is that instead of a laser, DLP 3D printers use a digital LED projector, which illuminates the entire layer simultaneously and not gradually like a laser in stereolithography does [18, 23]. It is assumed that DLP printing allow reproducing objects faster. However, this difference is not so great as to replace SLA 3D printers from the market of 3D printing.

As in SLA technology, there are two variants of devices for DLP 3D printing: in one the construction of the object occurs from the bottom-up (the operating platform is lowered), and in the other on the contrary — the construction of the object occurs from the top-down (the operating platform rises).

The main advantages of DLP 3D printing: higher printing speed as compared to SLA 3D printers; high printing accuracy; large selection of consumables; affordable cost of equipment due to the use of DLP projectors in printers, which are much cheaper than laser systems (as in SLA).

The main disadvantages of the technology: accuracy of printing is inferior to the accuracy of SLA 3D printing and depends not only on the 3D printer, but also on the used material (the more it is filled with pigments and light blockers, the more accurate products will be printed from it) and the environment (during polymerization namely in DLP printers a lot of heat is released, which leads to the acceleration of chemical reactions); the need for additional irradiation of the product after printing for final solidification.

Multi-jet 3D printing (polyjet and MJM 3D printing). 3D printing using PolyJet and MJM technology is similar to printing with a conventional inkjet 2D printer, but instead of using ink, layer-by-layer spraying of liquid light-sensitive polymer materials takes place on a special inner pad with a subsequent irradiation of each layer with ultraviolet light [19, 23]. MJM and PolyJet technologies are almost indistinguishable from each other. The difference in names is predetermined by the corresponding patents: Multi Jet Modeling technology belongs to the Company 3D Systems, and PolyJet — to the competing Company Stratasys.

With the help of a printhead equipped with nozzles, from which liquid consumables: the model material — material A and the support material — material B (Figure 3) are sprayed in thin layers in accordance with the computer CAD 3D model. Each layer is polymerized by the light of an ultraviolet lamp immediately after deposition. The layers are deposited to each other to create the specified three-dimensional model. As a result, a full-color three-dimensional object is produced that can be used immediately after the printing process is completed without additional surface treatment. The material of the supporting structures for the overhanged elements of the future model, used in the printing process, is easily removed after printing mechanically or washed off with water or dissolved in a special solution (for PolyJet 3D printing) or melted in a special furnace (for MJM 3D printing).

The main advantages of multi-jet 3D printing (PolyJet and MJM) 3D are high accuracy (0.016–0.085 mm) and speed of building models with a complex geometry, excellent physical and mechanical properties of products; easy removal of supports, absence of contact with liquid photopolymer and simple replacement of heads; possibility of surface treatment (gluing, grinding, painting, priming, deposition of various coatings, etc.); ability to print with different materials during manufacture of a single 3D model in a single working cycle; possibility of application in the office; high speed of manufacturing of large-scale projects.

The main disadvantage of the technology is the high cost of 3D printing.

SLS 3D printing. Selective laser sintering is one of the methods of 3D printing, which is widely used in expensive professional 3D printers and is characterized by high quality of products [21]. With its help it is possible to achieve the result close to the reproduction of products by injection molding. This procedure allows creating fully finished products of complex geometric shapes within a matter of hours, which explains its popularity among industrial organizations around the world.

The process of SLS printing consists in layer-by-layer sintering of powder material particles to form a physical object according to the specified CAD model. Sintering of the material occurs under the influence of a beam of one or more lasers (Figure 4).

Before the start of the construction process, a special section of the 3D printer is filled with consumables, after which the process of 3D printing is started. It is interesting that immediately before reproduction, the consumables are heated almost to the melting point, which facilitates and accelerates the operation of the SLS installation.

With the help of a laser installation and a scanning mirror, the laser beam is directed to the necessary areas of powder, sintering them together. After sintering the first layer, a special mechanism adds a thin layer of powder on the top of it, and the process takes place again until a complete construction of the product. In the process of printing, the platform of the operating chamber is constantly lowered (the step is equal to the thickness of the printed layer). Thus, the area of interaction between the material and the laser beam is always on the same level, and the reproduction of the object occurs from the bottom-up.

The SLS process does not require a construction of special support structures. Here, in capacity of supporting structures for construction a model of a complex geometry an unused powder is used, which after removal of the finished product is cleaned and can be reused for printing.

The main advantages of SLS 3D printing technology are the ability of creating complex models of high strength and quality (almost without visible layer-by-layer structure on the models); no need in constructing supports; high speed and efficiency of printing: SLS printers do not require a full melting of particles of the material that allows them to work much faster than other powder 3D printers; the process is almost waste-free — unused material (up to 90 % of powder) can be reused for printing.

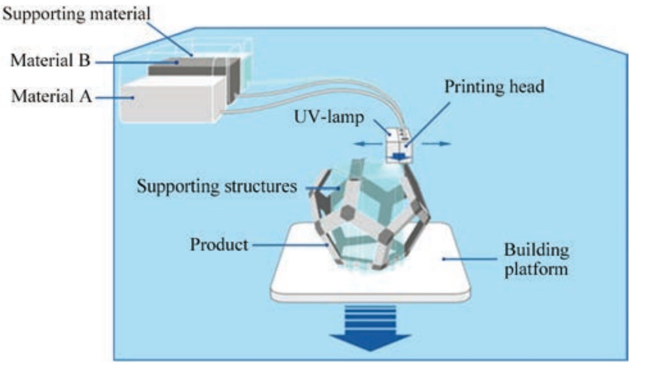

Figure 3. Scheme of PolyJet 3D printing process [20]

The main disadvantages of the technology are the complexity, cumbersomeness and a high cost of equipment; possible contamination during manufacturing: the powder is volatile and in case of careless handling it rises into the air, clogging the surrounding space and getting into the human lungs; it requires a long-term preheating of the powder, as well as time for cooling the finished model before removing the powder remnants; the complexity of processing the product after printing (annealing) in the special furnace for final sintering of the powder; shrinkage of a part after annealing sometimes reaches the values of 30 % (and on average 8−10 %) from the initial volume.

SHS 3D printing. Selective heat sintering is a 3D printing technology that is similar to selective laser sintering. The only significant difference is the use of an infrared radiation source instead of a laser for melting layers of thermoplastic powder [19, 23].

The process of SHS 3D printing is as follows: onto a plate made of quartz glass (mask), using a special material that reflects infrared radiation, a pattern is applied corresponding to the inverted cross-section of the current layer of the object. Then, the required amount of consumables for construction of one layer is supplied to the operating platform. The powder is spread with a special roller, and its excess is removed.

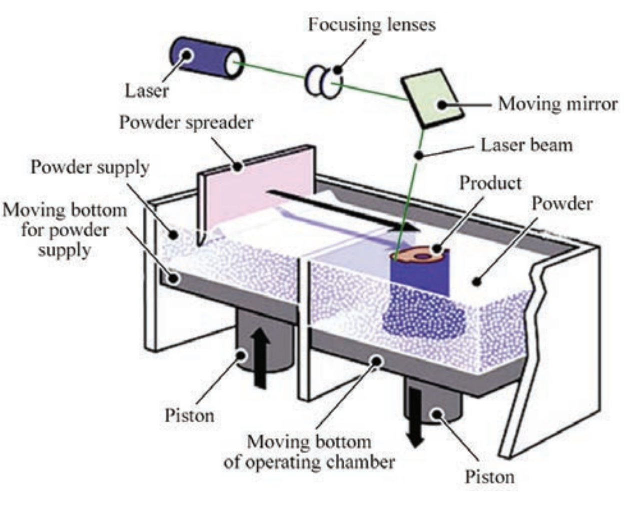

Figure 4. Scheme of SLS 3D printing process [22]

The quartz plate is located between the construction area and the radiation source. Once everything is ready, the sintering of the first layer begins. As far as the radiation goes through the plate, those its parts that reflect the radiation do not leave a trace on the operating platform. The remaining parts of the powder are sintered between each other. Upon completion of the first layer, the operating platform is lowered to the level of one layer, then the next portion of powder is deposited, the old pattern of the quartz plate is removed and a new one is deposited. This is how the whole model is built. After the end of 3D printing, a product is removed from the 3D printer, it is removed from the platform and cleaned of excess material. If necessary, the further processing is performed.

The main advantages of SHS 3D printing technology are high printing speed; the cost of SHS of the 3D printer and the cost of 3D printing on it is many times lower as compared to SLS with 3D printers; no need in construction of supports; the material left after printing can be reused, which reduces the cost of the process; the ability to create products with a complex geometry.

The main disadvantages of the technology are a low power efficiency, due to which the choice of consumables in this technology is much smaller; the need in additional annealing of the obtained products to achieve a higher strength; a lower strength of finished products as compared to the products made applying other industrial technologies.

CJP 3D printing. Color inkjet 3D printing allows a quick creation of both monochrome and colored objects from a composite powder [23]. As consumables two types of material are used — basic and binding. The basic material is powder plastic, which is used to form layers of the product, while the binder in the form of an adhesive substance simultaneously glues together the particles of the material in the required places in accordance with the computer 3D model and paints them in a preliminary specified color. First, the polymer powder is uniformly distributed in a thin layer over the entire platform plane of the operating chamber of the 3D printer (Figure 5), then a binder is applied to it and the platform is shifted down to the thickness of the layer (in the range from 0.089 to 0.102 mm). Then the process is repeated and as a result of subsequent cycles the finished product is formed layer-by-layer.

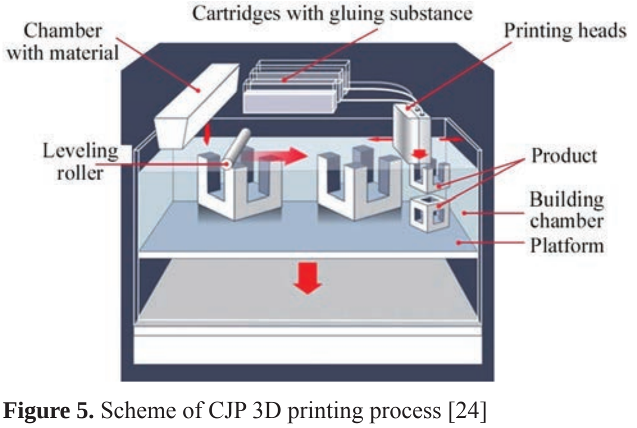

Figure 5. Scheme of CJP 3D printing process [24]

The powder, which is not used during the printing process to form a model (does not glue together), acts as a supporting structure, which allows creating objects with a complex geometry. At the end of 3D printing cycle, the same residual powder can be collected and reused. After the formation of a product is completed, a polymer powder remains on its surface, for the removal of which a cleaning chamber is provided in the 3D printer, which operates with the help of a compressed air. Then, the formed product is impregnated with cyanoacrylate, better known as «superglue» to increase strength and long life, as well as to achieve brighter colors.

The main advantages of the technology SHS 3D printing are low cost of manufacturing prototype due to a low cost of the material and its waste-free use; high speed of 3D printing and quality of models; available color palette reaches 390000 shades; the accuracy of construction surfaces with various degrees of complexity ranges from 0.4 to 0.1 mm; the wall thickness ranges from 0.5 mm, the layers are in the range from 0.089 to 0.102 mm; the prototypes are easily polished, glued and painted; lack of supporting structures.

The main disadvantages of the technology are that the models have a hygroscopic surface of high roughness and products require a careful processing after molding due to their brittleness.

LOM 3D printing. Manufacturing of three-dimensional objects by the method of lamination consists in layer-by-layer gluing of materials in the specified coordinates (from a computer 3D-model) with the subsequent cutting of remainders [23, 25]. As the consumables usually polyvinylchloride (PVC) film is used with a thickness of 0.15 mm in five colors: semi-transparent (amber), red, blue, cream, black. The product is formed on a special substrate of a movable (up and down) platform, which is supplied with consumables from a roll or in a separate sheet, distributed under a certain pressure and heated to the desired temperature by a roller (Figure 6). After that, the contour of the first layer of the model is cut with a laser. After that, the platform is lowered to the height of one layer, the roll is rotated (or a new sheet is applied) and the next layer of material is applied on the top of the first layer, on the lower part of which the binder is usually applied. Due to heating and depositing under pressure, a gluing (lamination) of layers occurs. The process of cutting layers and applying new sheets of material is repeated until the complete formation of a final product.

At the end of the 3D printing process, a part is removed from the 3D printer and cleaned of scraps. Then, if necessary, additional mechanical treatment: grinding, varnishing, painting is carried out.

The main advantages of LOM 3D printing technology are the possibility of full-color printing with a high resolution along the *X* and *Y* axes; availability and cheapness of consumables; for models with overhanging or horizontally protruding elements the formation of supporting structures is not required.

The main disadvantages of the technology are the extremely limited choice of materials for creating models; insufficiently high strength of products in the plane of layers — there is a risk of delamination; increased surface roughness; the thickness of the layer depends entirely on the thickness of the used sheet material, due to which the model is formed rough, and mechanical treatment for smoothing is not always available, as it can lead to delamination; a large amount of consumables (trimmings) goes to waste; manufacturing with increased fire hazard and smoke emission.

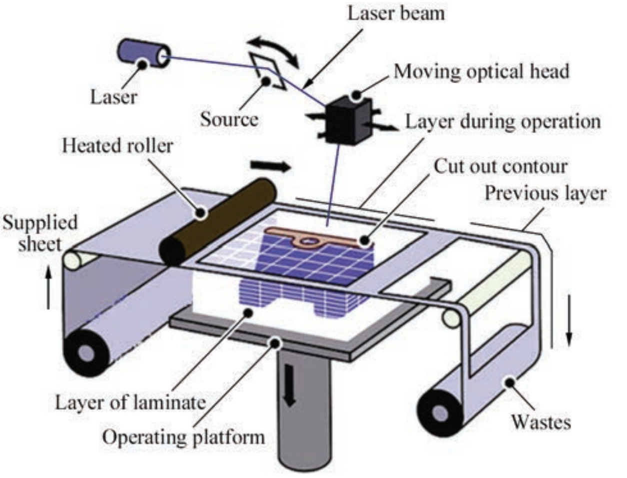

Figure 6. Scheme of LOM 3D printing process [26]

Conclusion

According to analysts, the prospects for today are such that in the near future, three-dimensional printing will be used in all areas of human activity [27]. Therefore, it is not surprising that the pace of development of additive technologies is rapidly increasing every year and today in the dynamics of development the market of additive technologies is ahead of other industries.

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