Management and Production Engineering Review
Volume 11 • Number 3 • September 2020 • pp. 65–73
DOI: 10.24425/mper.2020.134933

LITERATURE REVIEW ON EMISSIONS FROM ADDITIVE MANUFACTURING BY FDM METHOD AND THEIR IMPACT ON HUMAN HEALTH

Anna Karwasz, Filip Osiński

Poznań University of Technology, Poznań, Poland

Corresponding author:
Filip Osiński
Poznań University of Technology
Institute of Materials Technology
pl. M. Skłodowskiej-Curie 5, 60-965 Poznań, Poland
phone: +48 61 6475990
e-mail: filip.osinski@put.poznan.pl

Received: 31 January 2020
Accepted: 2 September 2020

Abstract
Additive manufacturing in recent years has become one of the fastest growing technologies. The increasing availability of 3D printing devices means that every year more and more devices of this type are found in the homes of ordinary people. Unfortunately, air pollution is formed during the process. Their main types include Ultra Fine Particles (UFP) and Volatile Compounds (VOC). In the event of air flow restriction, these substances can accumulate in the room and then enter the organisms of people staying there. The article presents the main substances that have been identified in various studies available in literature. Health aspects and potential threats related to inhalation of substances contained in dusts and gases generated during the process are shown, taking into account the division into individual types of printing materials. The article also presents the differences between the research results for 3d printing from individual plastics among different authors and describes possible causes of discrepancies.

Keywords
FDM, 3d printing, emissions, pollution, environment, health.

Introduction

3D printing is becoming more and more popular and available. It is used in many areas of life. From prototypes before mass production, to final products such as toys, prostheses of human limbs, chocolate or pizza. The size of manufactured parts or whole finished products does not matter. Even very small parts or buildings measuring several meters can be printed with additive manufacturing technology.

The advantages of 3D printing is the low cost of preparation of a CAD model and its manufacturing in unit or small amount production. 3D printing is an alternative to injection moulding, which is characterized by a high cost of unit or small series production, for example in the foundry industry [1]. The rapid development of incremental technologies has contributed to the fall in printer prices and increased their availability [1–3]. Everyone can afford to buy a small 3D printer for domestic needs. Another advantage of 3D printing is its short production time [3]. Just a few hours is enough to print the finished model along with its final processing. 3D prints have recently been used in medicine to diagnose cancer of various internal organs, its location, shape or size. In addition, if you have a model of the internal organ with cancer cells (tumor), you can practice its removal or practice sewing internal organs [1, 3–6]. There are also no contraindications to create complicated shapes, such as the internal organs of the human for example kidneys with supply channels [2–4]. Data from magnetic resonance imaging or computed tomography can be processed into a digital 3D model and then converted to STL printed for example from silicone. In this way, we can individually adjust the shape of a given internal organ that we will operate or adjust, as for example in the case of orthoses fitted to the hands or legs of a particular patient [2–6].

The additive manufacturing market has been developing very dynamically in recent years. The rapid growth is due to both the increased availability of printing devices and the continuous emergence of new areas in which it is possible to use 3D printing. It is estimated that in the years 2014–2017, the market value increased nearly 3 times from $ 3.07 billion to $ 8,68 billion [7]. It is expected that the following years will bring further market development and an increase in its value to nearly $ 52 billion [8]. Apart from the increase in the market value itself, it should be remembered that this technology will become more common and more and more often used in homes. The result is that all risks to the health of users of this type of printers should be recognized, and the problems that could have a significant impact on health should be eliminated in advance [9].

PM10 and PM2.5 suspended dust is the air pollution that causes the greatest damage to human health. The suspended dust is so light that it can float in the air. Some of its particles are so small (one thirty to one fifth the diameter of a human hair) that they not only penetrate deep into lungs, but also enter the bloodstream, as does oxygen [10]. These dusts fractions can consist of various chemical compounds, and their impact on our health and the environment depends on their composition. Some heavy metals, such as arsenic, cadmium, mercury and nickel, can also be found in suspended dust.

Fuel burning is the basic way of producing energy, as well for industry and household appliances. It is also one of the main sources of pollution of the atmosphere. It is important both due to the significant use of energy in the additive manufacturing process, as well as the most widely described emission sources in the literature. For example, to standardize the total emissions generated in technological processes, the CO_2-eq unit is used, which serves as a comparative measure for various processes or products in terms of air pollution. This measure was chosen precisely because CO_2 is the basic product of fossil fuel combustion. Taking into account the need to systematize certain emissions from 3d printing, refer to the emissions from combustion is compulsory. For example, the value of the CO2-eq coefficient for each kWh of electricity consumed in Poland is 0,836 kg CO2 [11]. Our cars, trucks, power plants and other industrial plants need energy. Almost all vehicles and plants use some kind of fuel and burn it to get energy. Fuel combustion usually changes the form of many substances, including nitrogen – the most common gas in our atmosphere. When nitrogen reacts with oxygen, nitrogen oxides and dioxides are formed in the air. When nitrogen reacts with hydrogen atoms, ammonia is formed, which is another air pollutant that has serious negative effects on human health and the environment. In practice, combustion processes release many other air pollutants, from sulfur dioxide and benzene to carbon monoxide and heavy metals. Some of these pollutants have a short-term effect on human health. Others, such as some heavy metals and persistent organic pollutants, accumulate in the environment. Thanks to this, they get into our food chain and, as a result, reach our table. Other impurities, such as benzene, can destroy the genetic material of cells and cause cancer in the event of prolonged exposure. Because benzene is used as an additive to gasoline, about 80% of benzene released into the atmosphere in Europe comes from the combustion of fuel used by vehicles. Other known carcinogenic contamination, benzo(a)pyrene, is released mainly through the combustion of wood or coal in home furnaces. Car fumes, primarily from diesel engines, are another source of benzo(a)pyrene. Itis not only carcinogenic compound, but it can also irritate the eyes, nose, throat and bronchi. It can be found in suspended dust.

Impact of air pollution on people

Every year, air pollution is the cause of health and life loss of many people. In Poland, about 19–22 thousand people over 30 untimely die each year due to the so-called "low emission" [12]. High concentrations of PM2.5, PM10 dust is harmful to health. It causes, among others, mucous membrane irritation, irritation of the eyes, nose, throat, dizziness, blurred vision. Sulfur dioxide pollutions causes headaches and anxiety. Pollution of air with ozone, dust, nitrogen oxides, sulfur dioxide or benzo (a)pyrene causes eye, nose and throat irritation and breathing problems. Air pollution with suspended dust, ozone or sulfur dioxide causes cardiovascular diseases. Air pollution with suspended dust causes also respiratory tract irritation, inflammation and infections. It also causes asthma and reduced lung efficiency, which can develop into a chronic obstructive pulmonary disease, and has a negative effect on the reproductive system. Particulate matter and benzo(a)pyrene cause lung cancer. Nitrogen dioxide has a negative effect on the liver, spleen and blood [8A]. Prolonged exposure to particulate matter increases the risk of developing cardiovascular and respiratory diseases, including lung cancer.

The size of the solid particles present in the inhaled air defines their place of accumulation in the human body. Particles with a diameter exceeding 10 µm (so called PM10) are separated in the throat and trachea area. Particles larger than 0.2 µm (200 nm) are already reaching the bronchi. On the other hand, the greatest threat to health are particles with dimensions around 100 nm, which enter the alveoli where they can be absorbed directly into the blood. The distribution of particulate matter in the human respiratory tract is described in Table 1.

Table 1
Entry of solid particles to human body according to the Weibul-Lung model [12].

Organ	Entry of solid particles	
	Amount of particles	Surface concentration [1/cm^2]
Trachea	1	2.54
Bronchi	2	2.33
Pulmonary bronchioles	4–6 × 10^4	2.13–180
Alveoli	5 × 10^5	10^3
Vesicular pouches	8 × 10^6	10^4

The costs associated with the loss of health and life caused by air pollution in Poland are very high and range between EUR 12.8 billion and EUR 30 billion annually [13]. If calculated per capita for every person aged over 30, the cost in 2016 was valued between 300 and 800 EUR.

Emissions from FDM process

Emissions of solid compounds

Consumer-grade 3D printers have gained popularity in recent years and become more and more popular in home use. Unfortunately the particles and gasses emitted from such devices can negatively affect indoor air quality and can potentially harm health, affecting the upper respiratory tract [14]. As part of many years of research, researchers collected particles emitted from 3D printers and conducted several tests to assess their effects on human health [14]. Studies have shown a toxic reaction to particles from different types of filaments used in 3D printers. Researchers took a closer look at the chemical composition of the particles and their potential toxicity. During printing, the heated filament releases volatile organic compounds (VOCs) that are emitted into the air near the printer and the printed object.

Model of formation and transformation of solids in the nozzle area is shown in Fig. 1. Particles are formed as a result of chemical changes caused by high temperature and pressure in the extruder such as:

- thermal depolymerization of the filament material,
- breakdown of chemical compounds,
- evaporation of compounds added to the material (stabilizers, antioxidants, dyes, etc.),
- coagulation of smaller particles (including VOCs).

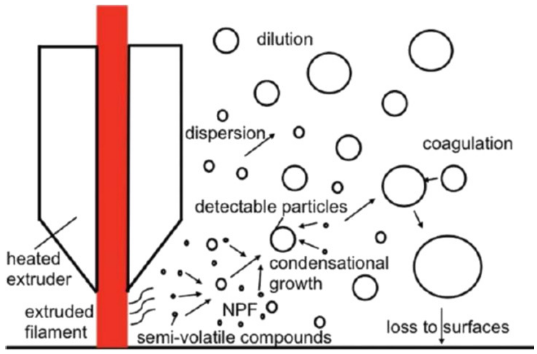

Fig. 1. Schematic of particle formation, growth, and loss processes [14].

In earlier studies, the team of researchers found that the higher temperature is required to melt the filament, the concentrations of the emitted compounds is higher [15]. As a‘ result, plastic fibers of acrylonitrile-butadiene-styrene (ABS), which require a higher temperature to melt (250–280°C), produced more emissions than fibers made of polylactic acid (PLA, 200–220°C), which melt at a lower temperature. To test the effect of emissions on live cells, scientists collaborated with the Weizmann Institute of Science in Israel. The researchers exposed human respiratory and rat immune cells to the concentration of particles from printers. They found that both ABS and PLA particles negatively affected cell viability, the latter provoking a more toxic response. Researchers also carried out chemical analysis of the particles to get a better insight into their toxicity and to allow comparison with the toxicity of particles found in urban environments (outdoors). Analysis – called oxidative potential – simulates the toxic reaction that an aerosol would have on cellular organisms. Toxicity tests showed that PLA particles were more toxic than ABS particles, but because printers emitted a lot more ABS – it is ABS emissions that are more disturbing. Tests indicate that exposure to these filament particles can over time be as toxic as air in an urban environment polluted by car emissions [15, 16].

The average size of particles produced during the process may also be a concern. For the vast majority of the filament materials, the size of the emitted solid particles varies in the range of 50–200 nm, wich is shown on Fig. 2. This is an important fact due the possibility of penetration of this size particles directly into the alveoli and then into the cardiovascular system. Also for the most popular particle sensors or air quality sensors, this particle size does not allow for effective information about a dangerous concentration of pollutants. The most widely used solutions in Europe allow the identification of particles with a size of 1000 nm, and very rarely – particles with a size above 300 nm. At present, the authors have no knowledge of non-professional devices that allow monitoring the concentrations of smaller particles.

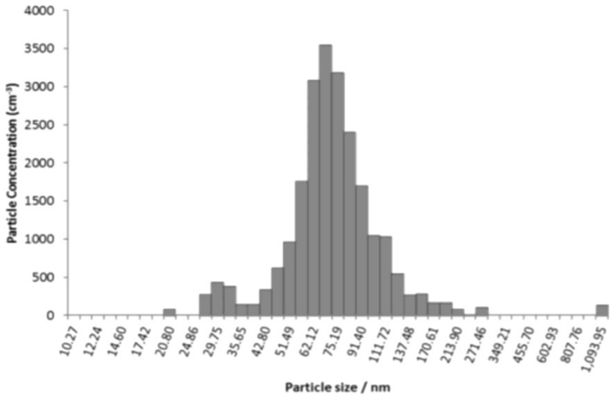

Fig. 2. Sizes of particles produced during the FDM process [17].

Studies by various authors have shown the presence of various compounds among the solid particles emitted from the 3D printing process. Examples of the most common compounds are presented in Table 2. Among all substances identified as components of dusts generated during additive manufacturing, Irganox 1076 reached the highest concentration for most of the tested filament materials. This substance, added as an antioxidant to plastics, especially those based on styrene polymers, can cause numerous health effects. The most important ones include skin, eye and respiratory tract irritation, but in long term can cause specific organ toxicity [20].

Another common compound in 3D printing dust is Bisphenol A (BPA). It is a plastic additive commonly used as a plasticizer and antioxidant. The most frequently observed health effects of exposure to BPA are: skin, eye and respiratory tract irritation, eye damage and fertility problems. Many studies also emphasize the carcinogenic properties of BPA, including an increased risk of breast and prostate cancer [21, 22].

Other compounds which constitute a relatively large share of dusts from the additive manufacturing process are: PAME, TPPO, TCEP. The first two compounds added as plasticizers can irritate eyes, mucous membranes and respiratory tract [H, I]. However, the greatest concern should be Tris-(2-chloroethyl)-phosphate (TCEP) which has found use as an anti-pyrogen in filaments. This substance, inhaled, can cause numerous health effects, including: degradation of immune system cells, kidney lesions, damage and loss of nerve cells. There are also known evidence of carcinogenicity of TCEP [23–27].

In the vast majority of cases, the solids that are released during additive manufacturing are approximately spherical in shape. Unfortunately, some researchers also identified rarer, but occurring, spindle-shaped particles. That shape of the particles additionally facilitates the penetration of mucous membranes and blood vessels, which translates into an increase in the penetration of certain substances into the body [29, 30].

Table 2
Table 2 Concentrations of organic compounds (ng/m^3) in chamber air collected on quartz fiber filter during 3D printing with several filaments [17–19].

Substance	PCABS Ivory	ABS Red	HIPS Red	PETG Black	ASA Blue
Caprolactam		39	–	–	59
4-tert-Butylphenol	30	–	–	–	33
2,4-Di-tert-butylphenol	17	–	–	6	28
Diethyl phthalate (DEP)	–	17	20	22	19
n-Octyl ether	19	41	19	–	
Lauryl acrylate	70	–	16	–	30
Tris-(2-chloroethyl)-phosphate (TCEP)	63	25	107	12	59
Diisobutyl phthalate (DiBP)	26	–	41	8	26
Methyl palmitate (PAME)	79	47	101	23	80
Di-n-butyl phthalate (DnBP)	17	10	24	–	18
Bisphenol A	143	185	–	–	424
Triphenylphosphine oxide (TPPO)	–	78	92	55	71
Tris(2,4-di-tert-butylphenyl) phosphate (TDTBP)	–	–	–	–	207
Irganox 1076	936	1043	–	–	603

The humidity of both the material and the air at the place of manufacture is not without significance for the process. In the case of moisture contained in the material itself, there are numerous problems with the correct plasticization of the filament. There are two main reasons for this. Firstly, a much higher specific heat capacity of water (4190 J/kgK) than the filment itself (range 900–2200 J/kgK depending on the material), which translates into insufficient or uneven heating of the material in the extruder. Secondly, the very high temperature expansion of water during boiling may lead to rapid pressure changes in the extruder. The changes in the filament caused by both phenomena and the reduction of the process stability may lead to more rapid changes on the surface of the molten material flowing from the extruder. The effect of such changes may be an increased emission of solid particles, by pulling microscopic particles of the material from its surface. Another element related to humidity is the moisture content in the air at the site of the process. As in the case of moisture in the filament, the specific properties of water may cause changes in the course of the process during the solidification of the material after leaving the extruder. Firstly, high air humidity may accelerate the coagulation and condensation growth mostly for compounds volatile at higher temperatures. Secondly, the loss of particles on the surface of the molten material can be significantly reduced by accelerating its solidification, which is shown on Fig. 3 [31].

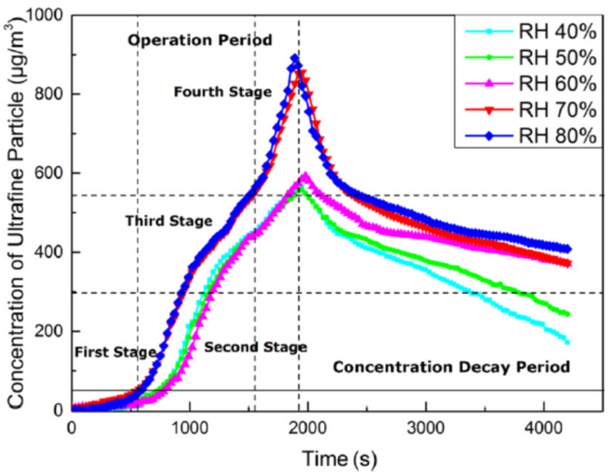

Fig. 3. PM2.5 concentrations under different humidity [31].

In addition, from the point of view of penetration of the particulate matter in the human body conditions (parameters of the air) in the printing room can be significantly affected. Relatively low air flow, required to maintain the appropriate quality of manufactured products, promotes the accumulation of particles in the room. Additionally, low air humidity and elevated temperature (especially when using more devices in one room) may cause drying out and micro-damage to the mucous membranes. This situation causes an additional impact of the process itself on the health of people staying for a long time in the printing rooms.

VOC emissions

Many people spend up to 90% of the day indoors – at home, work or school. The quality of the air people breathe in indoors also has a direct impact on their health. Although it may come as a surprise to many people, the air on a city street with moderate traffic can in fact be cleaner than the air in our houses or work. The results of the conducted tests indicate that the concentration of harmful air pollutants may be higher indoors than outside. In the past, indoor air pollution issues have received much less attention than outdoor air pollution problems, in particular pollution caused by industrial emissions and emissions generated by the transport sector. In recent years, however, the dangers of exposure to indoor air pollution have become more evident. Just imagine a freshly painted apartment with new furniture or a place of work, in which there is a heavy smell of cleaning preparations. The quality of the air in our homes, workplaces or public spaces varies greatly, depending on the material from which the object was built, the medium used to clean it, and the purpose of the room, as well as how it is used and installed ventilation system. Poor indoor air quality can be extremely harmful to vulnerable groups, such as children, the elderly and people with cardiovascular or chronic respiratory diseases such as asthma. The most important indoor air pollutants include radon (radioactive gas formed in soil), tobacco smoke, gases and particles formed as a product of fuel combustion, chemicals and allergens. Compounds such as carbon monoxide, nitrogen dioxide, particles and volatile organic compounds occur both indoors and outdoors [10].

In the course of various studies on emissions from 3D printing, about 216 different compounds that can be released in gaseous form during the manufacturing process have been identified. The averages and ranges of emissions of total volatile compunds (TVOC) from individual filament types are described in Table 3, while the comparison of the test results of different authors is presented separately for each filament type in Tables 4 to 6. Some of these substances are characteristic for the filament material. For example, styrene, formed during the degradation

Table 3
Average TVOC ERs and yields for different filament materials [28].

Emission	ABS	PLA	NYLON	HIPS
Average TVOC [ug/h]	835	193	1660	888
Range of values	506–1460	149–269	276–3050	–
Average TVOC [ug/g filament]	77	13	134	71
Range of values	37–116	9–18	20–249	–

Table 4
Results of various tests on VOC emissions from ABS [32–35].

ABS Emission	Aika 2019	Azimi 2016	Gu 2019	Stefaniak 2017
TVOC [ug/h]	835	1500–2340	918	683.6
Styrene [ug/h]	276	600–2100	384	251
Ethylbenzene [ug/h]	69.3	60–300	288	4.7–7.3
Benzaldehyde [ug/h]	71.5	–	48	–
Trichloroeten [ug/h]	–	–	6	–
Dodecanal [ug/h]	6.5	–	48	–
Acetaldehyde [ug/h]	53.6	–	–	7.7–16.3
Formaldehyde [ug/h]	24,7	–	–	–
1-butanol [ug/h]	19,8	–	72 (C-3)	–
Acethophenomene [ug/h]	63,1	–	12	–
p,m-Xylene [ug/h]	6.8	–	12	0.2–3.1
Ethanol [ug/h]	–	–	–	39.9–67.2
Acetone [ug/h]	–	–	–	15,0–62,4
Propylene glycol [ug/h]	–	0–60	–	–
Hexenal [ug/h]	–	0–60	–	–

Table 5
Results of various tests on VOC emissions from PLA [32, 33, 35].

PLA Emission	Aika 2019	Azimi 2016	Stefaniak 2017
TVOC [ug/h]	199.3	480-840	4176
Lacitide [ug/h]	111	300	–
Acetaldehyd [ug/h]	18.8	–	300
1-Butanol [ug/h]	17.8	–	–
Formaldehyd [ug/h]	7.0	–	–
Decanal [ug/h]	4.1	–	–
Benzaldehyd [ug/h]	4.1	–	–
Nonanal [ug/h]	2.9	60	–
Caprolactam [ug/h]	7.4	–	–
Styrene [ug/h]	1.6	48	–
Ethanol	–	120	4380

Table 6
Results of various tests on VOC emissions from PETG [34].

PETG Emission	Gu 2019
TVOC [ug/h]	78
Acetone [ug/h]	12
Acetic acid [ug/h]	24
n-Butyl actate [ug/h]	24
Benzaldehyd [ug/h]	12
Styrene [ug/h]	6

of ABS, is characteristic for working with this material. Other identified volatile substances may arise as a result of the evaporation of additives to the material, chemical transformations or the use of various types of adhesives on the working plate. According to the available studies, the TVOC emission range varies between 276 and 3050 µg. This amount may depend on many factors, such as:

- type of filament material,
- extruder temperature,

- use of adhesives,
- filament color,
- filament manufacturer (different producers ad different plasticizers, antioxidants and anti-pyrogens),
- humidity, both of the filament itself and the air in the process.

Among the research conducted so far in the field of VOC emissions in printing with ABS, there are significant differences in both the concentrations of the identified substances and the composition of individual VOC components. The results can be influenced by both the printing parameters, the research method or the device used (printer). However, the filament used in tests seems to have the greatest impact on results, as in the case of solid particles. Further research is needed, in which it will be possible to use filaments with a strictly defined content of dyes, antioxidants and other additives to compare how they affect the actual emission.

In the case of printing with ABS, the main volatile compound generated during printing is styrene. This compound is formed as a result of thermal depolymerization of ABS itself. According to most sources, the emission varies between 200–400 µg/h, but some studies indicate much higher values [32–35]. Styrene is considered a strong irritant to the eyes and mucous membranes. Numerous studies have shown that prolonged exposure to an environment with elevated styrene concentrations may lead to respiratory and heart diseases, neurodegenerative diseases and endocrine problems [36–38].

Another compound identified in virtually all studies about ABS printing is ethylbenzene. This compound is irritating to the eyes, may cause damage to the respiratory tract, and has a narcotic effect on the central nervous system [39]. Common compounds found in vapors from 3D printers include aldehydes such as formaldehyde, acetaldehyde and benzaldehyde. All substances that belong to this group are considered toxic and harmful to the respiratory apparatus. It is also worth paying attention to substances such as acetone or ethanol, which can be detected as process emissions and come from the residues after cleaning the print bed.

In addition to previously appearing substances, PLA printing is characterized by the appearance of Lacitide as the main constituent of VOC. This substance is the effect of the depolymerization of polylactic acid and may cause irritation of the eyes after prolonged stay in its vapors. It is also worth noting ethanol that appears in some studies – which is related primarily to the process of preparing the print bed for production.

VOC emissions resulting from 3D printing with PETG seem to be relatively poorly described in the literature. This may be largely related to the relatively low emission of volatile compounds compared to other materials commonly used in additive manufacturing. Such a situation may be related to the relatively high resistance of PETG to chemical transformations, high softening point or high viscosity during extrusion.

Conclusions

Despite relatively small amounts of gas and dust emissions during additive manufacturing, this topic cannot be ignored. Each of the emitted substances may pose a threat to health, especially during long exposure to it. In the case of limited air exchange, during the additive manufacturing process, both at home and in the case of farms, a significant increase in the concentration of pollutants in the air may occur. Additionally, the ability of some of the resulting substances to accumulate in the organisms of animals, means that constant stay in the room where 3D printing is carried out has a negative impact on human health.

The research on emissions from additive manufacturing carried out so far leads to the conclusion that this process may have a significant impact on the health of people staying in the printing rooms. Unfortunately, significant differences in the case of some studies do not facilitate the assessment of the exposure scale and the clear determination of the scale of the problem. The different research methods, taking into account various factors, and above all the use of different filaments with not fully described composition (plasticizers, dyes etc.), make it difficult to compare the results of individual tests.

Taking into account the rapid development of the FDM 3d printing market, it is reasonable to set clear boundaries in which the printing process is safe for health. This topic is discussed by scientists and individual users of 3d printers. Therefore, setting out simple rules, understandable for both professionals and amateurs, seems to be applicable in the future to reduce the occurrence of diseases resulting from air pollution.

The studies were carried out with a support from statutory activity financed by Polish Ministry of Science and Higher Education (0613/SBAD/8727).

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