ADRIAN DOMIŃSKI / ORCID: 0000-0002-7716-5597 / adominski@cmpw-pan.pl KHADAR DUALE / ORCID: 0000-0002-0497-2128 / kduale@cmpw-pan.pl TOMASZ KONIECZNY / ORCID: 0000-0001-8508-0422 / tkonieczny@cmpw-pan.pl PIOTR KURCOK / ORCID: 0000-0002-8748-1649 / pkurcok@cmpw-pan.pl MAREK KOWALCZUK / ORCID: 0000-0002-2877-7466 / mkowalczuk@cmpw-pan.pl JOANNA RYDZ / ORCID: 0000-0003-3972-7074 / jrydz@cmpw-pan.pl CENTRE OF POLYMER AND CARBON MATERIALS, POLISH ACADEMY OF SCIENCES, ZABRZE, POLAND ALEXANDER GRUNDMANN / ORCID: 0009-0005-7981-3796 / grundmann@comprisetec.de SIMON T. KAYSSER / ORCID: 0009-0000-1480-2504 / kaysser@comprisetec.de COMPRISETEC GMBH, RÖDINGSMARKT 20, 20459 HAMBURG, GERMANY

ADDITIVE MANUFACTURING TECHNOLOGY FOR MODERN PACKAGING

TECHNOLOGIA WYTWARZANIA PRZYROSTOWEGO W PRODUKCJI NOWOCZESNYCH OPAKOWAŃ

ABSTRACT: Appropriate packaging is essential to protect products from external contamination, physical damage or food spoilage. The latest innovations in the packaging industry are mainly limited to the development of new polymeric barrier materials and composite or green, environmentally friendly materials. However, recently, new active, and/or intelligent (smart) packaging is being developed that can extend the shelf life of a product, keep it in good condition and help control the quality of food products. This review presents the latest developments and applications of additive manufacturing in the production of smart food packaging.

Key words: intelligent/smart packaging, three-dimensional (3D) printing, additive manufacturing, material extrusion, polyhydroxyalkanoate, polylactide, polyester

STRESZCZENIE: Odpowiednie opakowanie jest niezbędne, aby chronić produkty przed zanieczyszczeniami z zewnątrz, uszkodzeniami fizycznymi lub zepsuciem się żywności. Najnowsze osiągnięcia w branży opakowań ograniczają się głównie do opracowania nowych polimerowych materiałów barierowych oraz kompozytowych lub ekologicznych materiałów przyjaznych dla środowiska. Jednak ostatnio opracowywane są nowe opakowania aktywne i/lub inteligentne (smart), które mogą wydłużyć okres przydatności do spożycia produktu, utrzymać go w dobrym stanie i pomóc kontrolować jakość produktów spożywczych. W niniejszym artykule przedstawiono najnowsze osiągnięcia i zastosowania wytwarzania przyrostowego w produkcji inteligentnych opakowań do żywności.

Słowa kluczowe: inteligentne opakowanie, druk trójwymiarowy (3D), produkcja przyrostowa, wytłaczanie materiału, polihydroksyalkanian, polilaktyd, poliester

1. INTRODUCTION

Appropriate packaging is essential to protect products from external contamination, physical damage, or food spoilage. Therefore, the packaging industry has been experiencing rapid growth. The observed technological progress in the packaging market is primarily due to lifestyle changes, rising consumer spending, and improving exports on a global scale. The requirements for modern packaging are determined by constant changes related to demographics, lifestyle, level of health safety, ecology and production environment, as well as the development of new markets [1]. Recently, innovations in the packaging industry have mainly been limited to the development of new barrier materials, i.e. new polymeric materials, composite materials, or new green and environmentally friendly materials [2]. However, recently, many scientists and manufacturers have met the expectations of consumers and developed new active, and/or intelligent (smart) packaging. This type of packaging can extend the product's shelf life, keep it in good condition, and help control the quality of food products. Intelligent packaging refers to packaging designs that use various sensors to provide information about the state of the contents, and active packaging maintains quality by adding, for example, antibacterial or antioxidant agents. Sensors, chemical or temperature indicators, gas emission indicators, and microbial growth indicators are all components of intelligent packaging solutions. The new antibacterial packaging, designed to improve food safety, is also interesting. This type of packaging extends the growth retardation of microorganisms or reduces the growth rate and final number of microorganisms [3]. There is a wide range of consumer opinions about intelligent and/or active food packaging. Smart package technology is perceived by consumers as a way to guarantee food quality and safety while also delivering real-time consumption information, minimising food loss, and decreasing food waste - a huge problem in the XXI century, taking into account the millions of tons of food that are wasted year [4]. However, consumers are worried about the expenses of this packaging, nevertheless. They have stated that they would accept a 10% increase in the product's overall cost as a reasonable margin [5]. On the one hand, smart packaging is used for food authentication (geographical origin, animal or plant species identification, production method, etc.), which would reassure customers that the products they get are from reliable companies. On the other hand, the food sector, however, is now generally unable to implement smart packaging since the existing technology employed would result in a significantly bigger rise in product costs than the 10% reported by consumers. Manufacturers and scientists are working to create fresh approaches to lower production costs. Currently, it seems that three-dimensional (3D) printing, or rather additive manufacturing, is a leading technology for creating packaging that meets consumer expectations. Although the additive manufacturing has been utilised in several industry sectors for about 20 years, interest in and application of the technology have significantly increased in the last years [6]. Additive manufacturing has ushered in a new era of innovation that has been spurred by its unique potential because of its availability, versatility, performance, and affordability. This has not gone unnoticed in the packaging industry. Additive manufacturing is gaining popularity due to its singular capacity to produce sophisticated designs that can be utilised for mass manufacturing while having a favourable influence on the economy and environment. This method permits changes in the composition and structure of the material through the printed product, modifying the physicochemical characteristics of the packaging adapted to the expected requirements. Innovative package designs have been produced using this technology, which includes die cuts and forms that are personalised smart packaging. Coaxial extrusion printing has been utilised to create smart food packaging systems that can track the quality of packed food and the environment in real-time to fulfil the rising consumer demand for safe food [7]. This review reports on the latest developments and applications of additive manufacturing in the production of smart food packaging to provide the reader with a comprehensive background for understanding the current state of knowledge.

2. ADDITIVE MANUFACTURING

Additive manufacturing has transformed the product creation process in numerous industries. In contrast to traditional processing methods such as subtractive manufacturing, injection moulding, extrusion or casting, additive manufacturing constructs objects layer by layer from a sliced digital model. Because of this distinctive process approach, additive manufacturing provides a manufacturing possibility with a high level of design flexibility. Conventional manufacturing techniques frequently limit the complexity of geometries, with internal ones often being impossible to manufacture. By using additive manufacturing, designers can produce intricate shapes as well as customised products that were previously unfeasible or economically impractical to manufacture. The complexity of components usually can be increased to a certain level with no influence on the production costs, depending on the chosen additive manufacturing process. This enables designers to incorporate even minor details into products without diminishing their complexity for the sake of lower production costs [8]. The ability to produce a wide variety of components without requiring extensive preparation beyond the computer-aided design (CAD) model, printable material, and the machine itself is the biggest advantage of additive manufacturing. In contrast, conventional manufacturing can be both time-consuming and costly to start production. For instance, in manufacturing, a single component may require special tools or moulds with lead times of several weeks. Additive manufacturing can eliminate such obstacles in many cases, enabling designers to obtain a physical object that corresponds to their CAD model within only a few hours of production time. Such a degree of flexibility allows for real ondemand and on-site manufacturing, which can be advantageous to the entire supply chain.

Due to these advantages, the use of additive manufacturing can be profitable from a purely economic point of view. This is particularly true for low-volume productions when it is not costeffective to invest in expensive mass production equipment such as moulds or tools. The literature provides several use cases in the industry, for example in the medical, dentistry, aerospace, and automotive industry [9,10]. In the context of packaging, additive manufacturing is experiencing an increase in attention. Its capability of high design flexibility leads to various possibilities, for example, the creation of individualised or intelligent packaging [11]. Despite these advantages, additive manufacturing may not be appropriate for all types of applications due to certain limitations in comparison to traditional manufacturing. These drawbacks should be taken into account when considering its usage. A major drawback is the limited range of available materials, which lag significantly behind those available for traditional manufacturing processes. There are different additive manufacturing processes, which are designed to work with specific materials. Material extrusion (commonly known as fused deposition modelling (FDM) trademarked by Stratasys Inc or fused filament fabrication (FFF) [12]) for example works with thermoplastic polymers, vat photopolymerisation (stereolithography) with UV-reactive resins that result in thermosets. In order to produce a wide range of materials, several machines using different manufacturing mechanisms are required [13].

Other factors determined by the additive manufacturing process are the mechanical properties as well as the production speed of products. In general, the time to produce a single part is significantly longer than using mass production methods such as injection moulding, which makes them not suitable for manufacturing large volumes of identical parts. Such mass production machines can produce thousands of parts in a very good as well as reproducible quality within a few minutes or even seconds per part. With additive manufacturing however, it usually takes hours to produce a component, while the manufacturing time per part usually cannot be reduced. On top of that, the manufacturing principle of the layer-by-layer build-up leads to significant anisotropic properties regarding the dimensional accuracy, mechanical properties as well as the surface quality [14]. Accordingly, the definition and introduction of standardised guality control, monitoring and assurance methods for additive manufacturing processes is a big challenge [8].

To sum up, additive manufacturing can offer advantages over traditional processing methods. The largest benefits are the design freedom and the possibility to manufacture final products, prototypes as well as tools on demand without any special preparation or expensive tooling. The customisability inherent in additive manufacturing, makes it a cost-effective manufacturing method for a lot of low production volume use cases.

The primary challenge to overcome is the implementation of standardised, reliable, and reproducible component quality, including suitable quality control and assurance techniques. These standards must be developed and adjusted over years of industrial use, regarding existing additive manufacturing processes and materials as well as the upcoming ones in this innovative field of the industry [15].

Material extrusion is one of the methods of additive manufacturing and a fast growing, relatively simple, technology for producing almost any 3D object of any shape, with relatively high resolution and low cost using CAD. Additive technologies are dynamically developing in many industries in prototyping processes, but also in the production of highly complex elements, small-batch production and in the field of innovative solutions to the problems and limitations of traditional technologies, but also increasingly in the case of personalised consumer products such as packaging or everyday items. The process involves creating components by adding building materials, usually layer by layer. The rapid development of additive manufacturing technology has enabled the development of the market for (bio)degradable polymer "inks" in the form of fibres (filament), granules, powders, solutions, and gels for the manufacture of specific products. Thermoplastics are the only group of plastics that can be processed by both extrusion and injection moulding. Above a certain temperature limit, thermoplastics undergo a plasticised state, in which they are capable of large deformations. This enables pressure moulding as well as additive manufacturing. The combination of additive manufacturing technology with (bio)degradable polymers and/or renewable raw materials gives almost unlimited application possibilities [16,17].

As additive manufacturing is increasingly used for personalised consumer goods and processing can affect mechanical and thermal properties, especially in the case of (bio)degradable polymers, the behaviour of printed materials under different environmental conditions is being studied. The extrusion process often causes a decrease in viscosity and a decrease in the average molar mass, which impairs the mechanical properties. Mixing time, temperature and drying also affect the degradation of polylactide (PLA)-based plastics [18]. Therefore, it is important to determine the influence of printing conditions and direction on the properties of the polymer. Research was carried out to find out the relationship between the processing conditions and the direction of additive manufacturing on the geometry of the element (printing orientation: orientation of the product relative to the printer's working platform, arrangement of the filament in accordance with the algorithm used - in the horizontal and vertical direction) and hydrolytic degradation tests to see how the printed objects behave during degradation. The usefulness of cosmetic applications of materials including dumbbell-shaped bars, and packaging prototypes obtained by additive manufacturing was also investigated [17].

Commercially available (bio)degradable filaments, often protected by patents, do not contain precise information about the composition, and therefore their response to unfavourable environmental factors (abiotic and biotic) is unspecified. Therefore, it is important to know the composition of the filament made of (bio)degradable polymers and to predict the properties of printed objects in order to be able to accurately understand the impact of environmental conditions on the properties of finished products and match appropriate raw materials to specific applications. Ex-ante testing of polymeric materials to identify and minimise potential failures of new (bio)degradable polymer products before they emerge is extremely important [19].

Processing (additive manufacturing) causes not only an increase in the crystalline phase of the polymer after printing compared to the initial filament, but also an increase related to contact with the heated printer build platform. After printing, a slight increase in glass transition temperature (T_g) was also observed due to an increase in ordering, which may also increase the stiffness of the material. Not only does the contact time with the 3D printer platform lead to an increase in the crystalline phase during printing, but also smaller surface areas of the dumbbell-shaped bars that accumulate more heat, resulting in an increase in the degree of ordering [11].

Similar relationships were observed in the case of printing prototypes of cosmetics packaging (container) from PLA/PHA (PHA – polyhydroxyalkanoate) and PLA. Its individual parts during material extrusion had a different time of contact with the printer's working platform. The bottom of the container and the lid had a longer contact time (15-18 min) and during this time they were subjected to increased temperature causing the growth of the crystalline phase in contrast to the walls set vertically to the platform (melting enthalpy (ΔH_m) and cold crystallisation enthalpy (ΔH_{cc}) were lower for the walls of the container). Therefore, the processing conditions, in particular the printing orientation of the individual parts of the container, influenced its properties, which may then affect the lifetime and degradation process of the entire container [20].

The use of (bio)degradable polymers, especially in medical applications, requires a proper understanding of their properties and behaviour in different environments. Components made of such polymers can be exposed to changing environmental conditions, which can cause defects. During standard tests of hydrolytic degradation, dumbbell-shaped bars made of PLA and PLA/PHA obtained by material extrusion technology showed an unexpected phenomenon of shrinkage, which was about 50% of the length of dumbbell-shaped bars, regardless of the printing direction. Typically, polymers such as PLA break down already in the initial stage of hydrolytic degradation (at 70°C after 7 days). However, in this case, no significant degradation occurred after 70 days of hydrolytic degradation at 70°C. The phenomenon of shrinkage during degradation resulted in the entrapment of amorphous oligomers and hydroxy acids in the polymer matrix. Low-molar-mass degradation products, due to lower water penetration into the matrix, remained in it, increasing the molar mass dispersion, while causing less disintegration of the dumbbell-shaped bars. The additional stress caused by cutting the dumbbell-shaped bars in half disturbed the degree of ordering of the polymer structure and led to a further increase in the molar mass distribution, which suggests that the cutting of both PLA and PLA/PHA dumbbell-shaped bars (both directions of printing) obtained by material extrusion technology in the vertical direction led to shrinkage compared to uncut, which further limited water penetration [21].

Determining the safe service life of products made of (bio)degradable polymers, and optimising and understanding the physico-chemical changes in their structure is crucial for their numerous applications. The continual development of new materials that are stronger, lighter or more versatile than previous ones must not only lead to improved safety, but also reduce environmental concerns as the complexity of recovering the value inherent in a used product increases. Today's product design challenges lead to the development of polymeric materials that are stable in use, yet susceptible to microbial attack during organic recycling. For each application of polymeric materials, understanding which polymeric materials are optimal for their target applications allows accurate prediction of behaviour and quality assessment throughout their lifecycle, under real-world conditions. Potential failures can be avoided if all factors are taken into account at an early stage of new product development, also obtained by material extrusion.

2.1 FILAMENTS FOR PACKAGING

There are a range of additive manufacturing techniques, such as material jetting and vat photopolymerisation but extrusion based additive manufacturing technology seem to be more appropriate for food packaging applications as this can be made from a variety of thermoplastic biocompatible materials. Some of the bio-based or (bio)degradable polymers that are used in additive manufacturing include, PLA, PHA, polycaprolactone (PCL), poly(butylene adipate-co-butylene terephthalate) (PBAT), poly(butylene succinate-co-butylene adipate) (PBSA) and poly(butylene succinate) (PBS) [22]. Natural polymers such as cellulose and chitosan are also suitable for additive manufacturing packaging applications. However, since these are not thermoplastic polymers and the heating of a filament is used in the extrusion-based additive manufacturing process, processing temperatures must be controlled to avoid degradation. In most cases these are mixed with other (bio)degradable polymers. PLA is among the most popular filament materials commonly used for material extrusion printers. The popularity of PLA is due to its bio-based origin, biocompatibility, and (bio)degradability as it is derived from renewable resources, such as corn starch or sugarcane, and therefore offers not only good degradability and biocompatibility, but also it has excellent printability and good dimensional accuracy and less prone to retraction [23]. Moreover, polymers like PLA can also be used to additive manufacture objects that will be in contact with food as it is approved by the US Food and Drug Administration (FDA) for food and pharmaceutical applications. PLA-based filaments have been used in additive manufacturing of intelligent food packaging applications [24]. PHA is a class of biodegradable and compostable polymers produced by bacteria during fermentation processes using renewable raw materials and is also used in additive manufacturing applications. In general, PHA can offer the possibility to modify their properties such as varying in toughness and flexibility depending on the specific combinations of different monomer units included in the polymer chain and therefore creating PHA based polymers with properties similar to other thermoplastics. PHA filaments can be degraded in both industrial composting and soil

environments. A common approach to modify and improve the inadequate properties of (bio)degradable polymers is to produce polymer blends. For instance, PLA is blended with PHA to produce a filament with improved mechanical properties and tailored biodegradability. Various filaments made from a combination of PLA and PHA are available on the market as a fully compostable alternative that can be home-composted [25]. PBAT is another biodegradable polymer that has been used for the production of filaments for additive manufacturing applications. PBAT is derived from fossil fuels but can undergo degradation in a composting environment. Films based on PBAT and silver containing nanoparticles were fabricated by additive manufacturing for antimicrobial food packaging applications [26]. Natural polymer such as chitosan has been reported for 3D-printed intelligent food packaging systems [27]. These additive manufactured chitosan-based films exhibit colour changes and were produced for both antibacterial and antioxidant properties, as a way of controlling the quality, freshness and preservation of cold meat. In another example, Zhou et al. reported an intelligent food packaging system based on cellulose nanofibers that was fabricated by coaxial additive manufacturing [28]. The system provides dual functions of both maintaining fruit freshness as well as visual monitoring.

Although the additive manufacturing process generates less plastic pollution compared to the conventional manufacturing processes, it nevertheless still is associated with a lot of plastic waste, both in the initial extrusion process to produce filaments, during printing, or failed prototypes, support materials and because most printed parts are single-use models that are discarded. Moreover, the availability of cheap consumer additive manufacturing has increased in the last 20 years, following the expiration of previous patents for additive manufacturing machines such as material extrusion technique. This leads to demands for the use of more environmentally friendly filaments that fit into the sustainability initiative's mantra of replacing petroleum-based raw materials with bio-based compostable filaments. There are efforts to tackle this issue by producing filaments from recycled plastics, especially from nonbiodegradable polymers, to reduce the consumption of virgin plastics and promote a circular economy [29]. In addition, this initiative contributes to reducing the waste and energy consumption associated with the production of new polymers. In this process, the polymer material is collected, shredded, remixed, and then homogenised back into reusable recycled filament. PLA is one of the polymers that are recycled [30]. Recycled PLA from food packaging and bottles is converted into filaments and is commercially available.

3. ACTIVE AND INTELLIGENT PACKAGING

Modern food packaging can be divided into (i) active packaging or (ii) intelligent packaging [31]. The term active packaging refers to packaging in which certain additives called "active compounds" are introduced directly into the packaging material, in a leaflet or on a label, or placed directly in the packaging to interact directly with the product and/or its environment to extend its shelf life. "Active compounds" contain active functional groups that, interact with food, slowing down its spoilage. Active packaging containing substances is most often used [32]: antioxidants, (i.e., plant or oil extracts, flavonoids or even green tea extracts); antimicrobials (i.e., nisin, chitosan, and propolis glycolic); carbon dioxide (CO₂) emitters (sodium bicarbonate, citric acid, and ferrous carbonate); oxygen (O_2) scavengers (the most commonly used group of O₂ absorbers are solutions based on the oxidation of iron or palladium, which are deposited in a sachet placed in the package) and ethylene scavengers (i.e. activated carbon, titanium dioxide, and potassium permanganate). Intelligent packaging, on the other hand, is defined as a solution based on the interaction between the packaging and the product (or its environment) to inform the consumer about the freshness of the product. Unlike active packaging, these intelligent packages are designed to detect and record information related to food quality [31]. It is extremely useful to increase the efficiency of information transfer throughout the product distribution chain and even in the consumers' homes by using intelligent temperature or pH sensors or radiofrequency identification (RFID) tags. The main purpose of freshness indicators is to signal when the quality of the packaged product is no longer acceptable. Changes in their appearance are usually consistent with chemical or microbiological changes to which the product is subjected.

Packaged products may release into the atmosphere e.g. CO₂, amines, or ethylene. They can also release microbial metabolites. The released substances react with the indicator and change its colour, which proves the freshness of the product. Freshness sensors based on colorimetric indicators, capable of changing colour by reacting with volatile compounds produced on packaged food, have become the simplest, most practical, and instrument-free solution that can detect the level of freshness of the packed product with the naked eye [33]. Similarly, the mechanism works for temperature indicators. Because temperature is one of the critical factors affecting the quality and safety of food products from the moment of production to final consumption. Temperature and time are important factors in the rate of microbial activity in food, often deviating from specifications during production, distribution, and storage. Time and temperature indicators can be used to assess the effectiveness of pasteurisation and sterilisation. As critical temperature ranges are established to control food quality, it is important to monitor the length of time food is held at or above the critical temperature. The temperature indicators integrated with intelligent packages monitor the temperature in or around food packages, as well as the time the food has been exposed to undesirable temperatures. These indicators display an irreversible visual signal. The indicators are in the form of labels or small devices made of interconnected foil layers that can be included in the package. Thanks to this, they can show the effect of time and temperature on the product, which allows for correlating potential changes in food quality with them. The scientific literature describes many examples of indicators sensitive to pH or temperature changes, prepared by immobilisation of natural food dyes obtained from various sources, such as: anthocyanins, curcumin, alizarin, shikonin, and betalains on polymeric materials. However, their applications on different food types, including meat, seafood, and dairy products are usually specific, e.g. anthocyanins are most often used to determine the freshness of pork or chicken. and alizarin is more often used to determine the freshness of fish [34]. Recently, in an innovative approach to food quality control, a tactile label that becomes rough when food is no longer fresh is an example of a non-colorimetric indicator of food freshness [5]. The discussion on modern packaging must include RFID, which is currently the most important technology. RFID is a technology that uses electromagnetic fields to automatically identify and track product data. An RFID tag is a small device that can be attached to an object so that the object can be identified and tracked [35]. The tag is composed of a microchip, an antenna, and a substrate or encapsulation material. The microchip stores data whereas the antenna transmits and receives the data. The microchip and antenna attached to the substrate are referred to as the inlay. The RFID device transmits digital data, often an inventory number used to identify the item, back to the reader when it receives an electromagnetic interrogation pulse. The chip modulates the waves that the device sends back to the reader which converts the new waves into digital data. The inventory of products or supply chain may be tracked using this number.

4. ADDITIVE MANUFACTURING IN MODERN PACKAGING

To monitor the internal atmosphere within a food package and assist in food quality control, a sensor with composite polymer filaments containing the O₂-sensitive luminophore platinum (II) tetrakis(pentafluorophenyl)porphyrin was recently developed [36]. The luminophore gradually changed from a bright red colour to a pale pink colour with increasing O_2 concentration. Silicon dioxide (SiO₂) nanoparticles were coated with the luminophore and subsequently mixed with either polyethylene (low-density polyethylene, LDPE) or PLA to create the composite filament that was used in the additive manufacturing process. A material extrusion 3D printer was then used to print a 3D array of O2-sensitive luminophore/SiO2/LDPE or luminophore/SiO₂/PLA dots on a poly(ethylene terephthalate) (PET). As these indicators monitor O2 concentration in the internal packaging atmosphere, they can be incorporated into all types of food packaging to indicate a breach in package integrity. To monitor the package integrity of bottled liquid food products, a 3D-printed temperature-sensitive conductive filament was incorporated into a PET bottle cap that was used to measure the open and closed states of a sealed bottle as well as the temperature. The cap was created out of an exterior

cap that made it possible to attach it to a bottle and a spring that kept the contacts and the filament-based sensor apart. An electrical circuit is made by connecting the top and bottom contacts to the top and bottom of the cap and the spring, respectively. The method was developed by utilising an additive manufacturing process. To create a conductive filament, powdered conductive graphite was combined with a PLA matrix. The rise in filament resistance with rising ambient temperature served as the foundation for the sensing mechanism. By closing the gap between the top and bottom contacts, the circuit was completed, and this relationship was used to determine whether the cap was in the open or closed. When the cap is closed, it registers a resistivity of between 20-40 °C, but when it is open, it only registers a resistance of at all temperatures. This cap is designed to analyse the open or closed condition of the cap to monitor the package integrity of temperature-sensitive liquid products, such as carton-packaged or bottled milk or drinks. Smartphones can connect to it to check the data that is stored there [24]. Another example of a proposed smart component created by additive manufacturing is an RFID sensor comprised of an antenna atop a liquid crystal elastomer, prepared from 1,4-bis-[4-(6-acryloyloxhexyloxy)benzoyloxy]-2-methylbenzene, n-butylamine and photoinitiator (I-369) temperature-sensitive array and a T-match impedance. This RFID device measures food quality by monitoring ambient temperature. A 3D printer was used to posit the temperature-sensitive liquid crystal elastomer into a substrate to create a flat element array above the sensor's ground plane. The elastomer array reversibly actuated the RFID in response to the changes in ambient temperature. As the ambient temperature rises, the flat liquid crystal elastomer elements become dome-shaped, resulting in an increase in the space between the RFID antenna and the ground plane. When the temperature is decreased, the array returns to its original flat configuration, resulting in a decrease in the distance between the RFID antenna and the ground plane. Any temperature above critical causes the RFID change in operating frequency, which is communicated to nearby reader devices. This simple RFID device can be attached to packaged foods that pass through the cold supply chain, i.e., frozen products, dairy products, or eggs [37]. Additive manufacturing offers extraordinary opportunities in the design of complex devices. Recently scientists used additive manufacturing technology to print a colorimetric sensor for detecting Escherichia coli 0157:H7 [38]. The developed biosensor was based on gold nanoparticles, which were able to determine the concentration of *E. coli* bacteria using the colour change of the nanoparticles. A 3D printer was used to create a mould for manufacturing a microfluidic device consisting of two mixing channels, a separation chamber and a detection chamber. The biosensor was made by mixing nanoparticles, catalase and antibodies capable of detecting bacteria in the mixing channel of a microfluidic device to form an AuNP-bacteria-PS complex (AuNP - gold nanoparticle, PS - polystyrene). A cross-linking agent (mixture of hydrogen peroxide (H_2O_2) , tyramine and horseradish peroxidase) was added to the obtained complex, which changed the colour of the complex from red to blue. Then, the mixture obtained in this way was introduced into the detection chamber (package), where, with the increase in the concentration of E. coli bacteria, the colour of the gold nanoparticle complex changed from blue to red. Depending on the intensity of the colour, the concentration of the target bacteria is determined. Additive manufacturing has also been used to construct a biosensor for detecting the bacterium causing gastroenteritis - Salmonella typhimurium [39]. The 3D-printed biosensor consisted of a filter that prevented the device from being blocked by large particles and allowed the unhindered passage of bacteria to the second part - a linear microchannel for observing and counting fluorescent spots. The biosensor used a fluoroimmunoassay, a type of enzyme immunoassay that uses fluorogenic markers. S. typhimurium antibodies and iron-based magnetic nanoparticles were used to separate the target bacteria by forming magnetic nanoparticle (MNP)-bacteria complexes. These complexes then reacted with fluorescent microspheres to form fluorescent MNP-bacteria complexes that were injected into the microchannel of the printed devices. This biosensor worked with a smartphone to detect the target bacterium online. The light from the smartphone served as the excitation source, while a video processing application monitored the fluorescence

intensity moving along the microchannel and calculated the local concentrations of *S. typhimurium* in real time. Vat photopolymerisation (stereolithography) was also used to create a temperature-sensitive CO₂ sensor by in situ optical printing of photocrosslinkable poly(1-allyl-3-vinyl imidazolium bromide) with a high number of imidazolium functional groups. The stereolithography technique was chosen because of the extreme precision of printing, thanks to which it was possible to print the ultra-miniature sensors of Fabry-Pérot polymer interferometers. This miniaturised sensor can remotely and simultaneously measure CO₂ concentrations and temperatures in very small spaces, making it promising for many applications, from off-gas detection to food quality control [40].

5. CONCLUSIONS AND FUTURE OUTLOOK

Additive manufacturing is playing an increasingly important role in creating smart packaging that is equipped with advanced features and interactive elements. This technology allows sensors, electronics, and other components to be integrated into the printing process itself, opening up new opportunities for manufacturers and consumers. Additive manufacturing allows the creation of smart packaging with built-in sensors that can monitor various parameters. For example, packages can be equipped with temperature, humidity, or force sensors. This data can be used to track the storage conditions of products to ensure their quality and safety. In the future additive manufacturing, could make it possible to fabricate packaging with interactive elements such as print buttons, touch screens, or other user interfaces without having to assemble additional parts. This allows for the creation of integrated packages, such as RFID, which enables communication with mobile devices or other systems. Smart packaging produced using additive manufacturing can also have the ability to track and monitor products in real time. Thanks to built-in identification technologies such as QR codes, barcodes, or RFID tags, manufacturers can track the origin, history, and authenticity of products. This can contribute to combating counterfeiting and ensuring transparency and trust for consumers. In addition, additive manufacturing enables the personalisation of smart packaging, which can be important in marketing and customer interaction. The manufacturer can create unique patterns, logos, or labels that will distinguish the product on the store shelf and attract the attention of customers. This individualisation of packaging can be especially important for brands that focus on uniqueness and differentiation in the market. So far, there are few reports on the use of additive manufacturing to create intelligent packaging, but it seems that in the next few years, technology may be leading in the market. Additive manufacturing technology is more energy efficient and minimises the amount of waste because only the material necessary to create the object is used. In addition, additive manufacturing enables the creation of packages from recycled materials or biodegradable plastics, which contributes to reducing the negative impact on the environment. All in all, additive manufacturing creates new opportunities for smart packaging. It allows the integration of sensors, electronics, and interactive elements in the printing process itself, which enables the creation of monitoring, interactive and personalised packaging. Smart packaging based on additive manufacturing can contribute to improving product quality and ensuring the safety, traceability, and authenticity of products.

REFERENCES

- J.W. Han, L. Ruiz-Garcia, J.P. Qian, X.T. Yang, Food Packaging: A Comprehensive Review and Future Trends, Compr Rev Food Sci Food Saf 17(4) (2018) 860-877.
- [2] S. Nida, J.A. Moses, C. Anandharamakrishnan, 3D printed food package casings from sugarcane bagasse: a waste valorization study, Biomass Conv Bioref (2021).
- [3] M. Vanderroost, P. Ragaert, F. Devlieghere, B. De Meulenaer, Intelligent food packaging: The next generation, Trends in Food Science & Technology 39(1) (2014) 47-62.
- [4] C. Chen, A. Chaudhary, A. Mathys, Nutritional and environmental losses embedded in global food waste, Resources, Conservation and Recycling 160 (2020) 104912.
- [5] C.T. Tracey, A.L. Predeina, E.F. Krivoshapkina, E. Kumacheva, A 3D printing approach to intelligent food packaging, Trends in Food Science & Technology 127 (2022) 87-98.
- [6] A. Jandyal, I. Chaturvedi, I. Wazir, A. Raina, M.I. Ul Haq, 3D printing A review of processes, materials and applications in industry 4.0, Sustainable Operations and Computers 3 (2022) 33-42.
- [7] X. Luo, Application of inkjet-printing technology in developing indicators/sensors for intelligent packaging systems, Current Opinion in Food Science 46 (2022) 100868.
- [8] T. Pereira, J. V. Kennedy, J. Potgieter, A comparison of traditional manufacturing vs additive manufacturing, the best method for the job, Procedia Manufacturing 30 (2019) 11-18.
- [9] M. Attaran, The rise of 3-D printing: The advantages of additive manufacturing over traditional manufacturing, Bus Horiz 60(5) (2017) 677-688.

- [10] N. Hopkinson, P. Dicknes, Analysis of rapid manufacturing using layer manufacturing processes for production, Proc Inst Mech Eng Part C: J Mech Eng Sci, 217(1) (2003)31-39.
- [11] J. Gonzalez Ausejo, J. Rydz, M. Musioł, W. Sikorska, H. Janeczek, M. Sobota, J. Włodarczyk, U. Szeluga, A. Hercog, M. Kowalczuk, Three-dimensional printing of PLA and PLA/PHA dumbbell-shaped specimens of crisscross and transverse patterns as promising materials in emerging application areas: Prediction study, Polym. Degrad. Stab. 156 (2018) 100-110.
- [12] ASTM ISO/ASTM52900-21: Additive manufacturing General principles Fundamentals and vocabulary, ISO/TC 261 Additive manufacturing (2021).
- [13] D. Bourell, J.P. Kruth, M. Leu, G. Levy, D. Rosen, A.M. Beese, A. Clare, Materials for additive manufacturing, CIRP Ann 66(2) (2017) 659-681.
- [14] J. Ghorbani, P. Koirala, Y.-L. Shen, M. Tehrani, Eliminating voids and reducing mechanical anisotropy in fused filament fabrication parts by adjusting the filament extrusion rate, J Manuf Process 80, (2022) 651-658.
- [15] B. T. Riecken, S. T. Kaysser, Z. Kállai, J. Karsten, J. Hoppe, T. Konieczny, M. Hoppe, A. Lühring, P. Bitomsky, C. A. Keun, T. Schüppstuhl, B. Fiedler, Minimizing mechanical anisotropy in fused filament fabrication through innovative thermoset materials and additive manufacturing process, In: SAMPE Seattle Conference Proceeding, United States, April 2023.
- [16] J. Włodarczyk, W. Sikorska, J. Rydz, B. Johnston, G. Jiang, I. Radecka, M. Kowalczuk, 3D processing of PHA containing (bio)degradable materials, Chapter 6, In: M. Koller (ed.), Current advances in biopolymer processing & characterization, Biomaterials – Properties, Production and devices series, Nova Science Publishers, New York, 2017, pp. 121-168.
- [17] W. Sikorska, M. Musioł, K. Duale, J. Rydz, B. Zawidlak-Węgrzyńska, Biodegradable polymers. Value chain in the circular economy, CRC Press, Boca Raton, London, New York 2024.
- [18] W. Sikorska, J. Richert, J. Rydz, M. Musioł, G. Adamus, H. Janeczek, M. Kowalczuk, Degradability studies of poly(L-lactide) after multireprocessing experiments in extruder, Polym Degrad Stabil 97 (2012) 1891-1897.
- [19] J. Gonzalez Ausejo, J. Rydz, M. Musioł, W. Sikorska, M. Sobota, K. Włodarczyk, G. Adamus, H. Janeczek, I. Kwiecień, A. Hercog, B. Johnston, H.R. Khan, V. Kannappan, K.R. Jones, M.R. Morris, G. Jiang, I. Radecka, M. Kowalczuk, A comparative study of three-dimensional printing directions: The degradation and toxicological profile of a PLA/PHA blend, Polym Degrad Stab 152 (2018) 191-20.
- [20] J. Rydz, W. Sikorska, M. Musioł, H. Janeczek, J. Włodarczyk, M. Misiurska-Marczak, J. Łęczycka, M. Kowalczuk, 3D-Printed polyester-based prototypes for cosmetic applications – Future directions at the forensic engineering of advanced polymeric materials, Materials 12(6), 994 (2019) 20 pages.
- [21] J. Rydz, J. Włodarczyk, J. Gonzalez Ausejo, M. Musioł, W. Sikorska, M. Sobota, A. Hercog, K. Duale, H. Janeczek, Three-dimensional printed PLA and PLA/PHA dumbbell-shaped specimens: Mater ial defects and their impact on degradation behavior, Materials 13(8) (2020) 2005, 16 pages.
- [22] E. Gkartzou, E.P. Koumoulos, C.A. Charitidis, Production and 3D printing processing of bio-based thermoplastic filament, Manufacturing Review 4 (2017) 1.
- [23] J. Milde, R. Hrusecky, R. Zaujec, L. Morovic, A. Gorog, Research of ABS and PLA materials in the process of fused deposition modeling method, In: Proceedings of the 28th International DAAAM Symposium 1 (2017) 0812-0820.
- [24] S.L. Marasso, M. Cocuzza, V. Bertana, F. Perrucci, A. Tommasi, S. Ferrero, L. Scaltrito, C.F. Pirri, PLA conductive filament for 3D printed smart sensing applications, Rapid Prototyping Journal 24(4) (2018) 739-743.

- [25] P.H.M. Cardoso, R.R.T.P. Coutinho, F.R. Drummond, M.d.N. da Conceição, R.M.d.S.M. Thiré, Evaluation of Printing parameters on porosity and mechanical properties of 3D printed PLA/PBAT blend parts, Macromolecular Symposia 394(1) (2020) 2000157.
- [26] M.C. Biswas, B.J. Tiimob, W. Abdela, S. Jeelani, V.K. Rangari, Nano silicacarbon-silver ternary hybrid induced antimicrobial composite films for food packaging application, Food Packaging and Shelf Life 19 (2019) 104-113.
- [27] S. Li, Y. Jiang, Y. Zhou, R. Li, Y. Jiang, M. Alomgir Hossen, J. Dai, W. Qin, Y. Liu, Facile fabrication of sandwich-like anthocyanin/chitosan/lemongrass essential oil films via 3D printing for intelligent evaluation of pork freshness, Food Chem 370 (2022) 131082.
- [28] W. Zhou, Z. Wu, F. Xie, S. Tang, J. Fang, X. Wang, 3D printed nanocellulosebased label for fruit freshness keeping and visual monitoring, Carbohydr Polym 273 (2021) 118545.
- [29] F.A. Cruz Sanchez, H. Boudaoud, S. Hoppe, M. Camargo, Polymer recycling in an open-source additive manufacturing context: Mechanical issues, Additive Manufacturing 17 (2017) 87-105.
- [30] K. Mikula, D. Skrzypczak, G. Izydorczyk, J. Warchol, K. Moustakas, K. Chojnacka, A. Witek-Krowiak, 3D printing filament as a second life of waste plastics-a review, Environ Sci Pollut Res Int 28(10) (2021) 12321-12333.
- [31] T. Janjarasskul, P. Suppakul, Active and intelligent packaging: The indication of quality and safety, Crit Rev Food Sci Nutr 58(5) (2018) 808-831.
- [32] C. Vilela, M. Kurek, Z. Hayouka, B. Röcker, S. Yildirim, M.D.C. Antunes, J. Nilsen-Nygaard, M.K. Pettersen, C.S.R. Freire, A concise guide to active agents for active food packaging, Trends in Food Science & Technology 80 (2018) 212-222.
- [33] H. Yousefi, H.M. Su, S.M. Imani, K. Alkhaldi, M.F. CD, T.F. Didar, Intelligent food packaging: A review of smart sensing technologies for monitoring food quality, ACS Sensors 4(4) (2019) 808-821.
- [34] R. Priyadarshi, P. Ezati, J.-W. Rhim, Recent advances in intelligent food packaging applications using natural food colorants, ACS Food Science & Technology 1(2) (2021) 124-138.
- [35] P. Kumar, H.W. Reinitz, J. Simunovic, K.P. Sandeep, P.D. Franzon, Overview of RFID technology and its applications in the food industry, J Food Sci 74(8) (2009) R101-6.
- [36] D. Yusufu, R. Han, A. Mills, 3D printed O2 indicators, Analyst 145(12) (2020) 4124-4129.
- [37] J.G. Y. Shafiq, S.V. Georgakopoulos, H. Kim, C.P. Ambulo, T.H. Ware, A novel passive RFID temperature sensor, 2018 IEEE International Symposium on Antennas and Propagation & USNC/URSI National Radio Science Meeting (2018) 1863-1864.
- [38] L. Zheng, G. Cai, S. Wang, M. Liao, Y. Li, J. Lin, A microfluidic colorimetric biosensor for rapid detection of Escherichia coli 0157:H7 using gold nanoparticle aggregation and smart phone imaging, Biosens Bioelectron 124-125 (2019) 143-149.
- [39] S. Wang, L. Zheng, G. Cai, N. Liu, M. Liao, Y. Li, X. Zhang, J. Lin, A microfluidic biosensor for online and sensitive detection of Salmonella typhimurium using fluorescence labeling and smartphone video processing, Biosens Bioelectron 140 (2019) 111333.
- [40] J. Wu, M.-j. Yin, K. Seefeldt, A. Dani, R. Guterman, J. Yuan, A.P. Zhang, H.-Y. Tam, In situ μ-printed optical fiber-tip CO₂ sensor using a photocrosslinkable poly(ionic liquid), Sensors and Actuators B: Chemical 259 (2018) 833-839.

Acknowledgment. This work was supported by the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 872152, project GREEN-MAP, an international project co-financed by the program of the Minister of Science and Higher Education entitled "PMW" in the years 2020–2023; contract No. 5092/H2020/2020/2