Kaunas University of Technology
Faculty of Mechanical Engineering and Design

Design of Plant-Based Biodegradable Plastic Composites for Sustainable Packaging

Master's Final Degree Project

Lina Stabingytė
Project author

Lect. Laura Gegeckienė
Supervisor

Kaunas, 2021

Kaunas University of Technology

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Industrial Engineering and Management (6211EX018)

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Supervisor

Lect. Ingrida Venytė
Reviewer

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Lina Stabingytė

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Task of the Master's final degree project

Given to the student – Lina Stabingytė

1. Title of the project

Design of Plant-Based Biodegradable Plastic Composites for Sustainable Packaging

(In English)

Augalinės kilmės biologiškai skaidžių plastiko kompozitų, skirtų tvarioms pakuotėms gaminti, kūrimas

(In Lithuanian)

2. Aim and tasks of the project

Aim: To design plant-based biodegradable plastic composites in the use of sustainable packaging.

Tasks:

1. Develop comparable plant-based biodegradable plastic composite films;
2. Compare mechanical properties of the films;
3. Investigate the technological process of the production.
4. Investigate further composting, biodegradation and recycling;

3. Initial data of the project

N/A

4. Main requirements and conditions

Environmental conditions: 21-22 °C, relative humidity: 45-55 %, distilled water, maize starch, potato starch, pea fibre, sorbitol, glycerol, acetic acid, sheets of glass 210×297 mm, *Zwick Z005* tensile testing machine, *Dino light* microscope, *Lonis Schopper Leipzig* automatic micrometre.

Project author	Lina Stabingytė		2021-03-08
	(Name, Surname)	*(Signature)*	*(Date)*
Supervisor	Laura Gegeckienė		2021-03-08
	(Name, Surname)	*(Signature)*	*(Date)*
Head of study field programs	Regita Bendikienė		2021-03-08
	(Name, Surname)	*(Signature)*	*(Date)*

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Study field and area (study field group): Production and Manufacturing Engineering (E10), Engineering Sciences (E).

Keywords: biodegradable, plastic, starch

Kaunas, 2021. 55 p.

Summary

The study aimed to produce and investigate biodegradable films of plant origin to assess the suitability for the production of sustainable packaging. In this work, films made of potato starch, maize starch, plasticizers – glycerin and sorbitol, pea fibres, vinegar, and distilled water were compared. To find out if pea fibres can reinforce the polymer structure, which starch, and which plasticizer is most suitable for making a strong film. Starch-derived polymers have poorer mechanical properties but very fast disintegration times. As the need for sustainability grows globally, this alternative is becoming necessary to consider for short shelf life and easy degradation of polymer packaging. In the production of comparable films, the films with potato starch and combined with a sorbitol plasticizer had the best mechanical properties. Pea fibres had a positive effect in enhancing the polymer structure but did not increase the direct film strength. In order to develop the production of biodegradable plastics, a balance needs to be struck between easily degradable, strength-enhancing, moisture-resistant materials.

Stabingytė Lina. Augalinės kilmės biologiškai skaidžių plastiko kompozitų, skirtų tvarioms pakuotėms gaminti, kūrimas. Magistro baigiamasis projektas, vadovė lekt. Laura Gegeckienė; Kauno technologijos universitetas, Mechanikos inžinerijos ir dizaino fakultetas.
Studijų kryptis ir sritis (studijų krypčių grupė): Gamybos inžinerija (E10), Inžinerijos mokslai (E).

Reikšminiai žodžiai: biologiškai skaidus, plastikas, krakmolas

Kaunas, 2021. 55 p.

Santrauka

Tyrimo tikslas pagaminti ir ištirti augalinės kilmės biologiškai skaidžias plėveles, siekiant įvertinti tinkamumą tvarios pakuotės gamybai. Šiame darbe tarpusavyje lygintos plėvelės iš bulvių krakmolo, kukurūzų krakmolo, plastifikatorių – glicerino ir sorbitolio, žirnių skaidulų, acto ir distiliuoto vandens. Siekta išsiaiškinti ar žirnių skaidulos gali sutvirtinti polimero struktūrą, kuris krakmolas ir kuris plastifikatorius yra tinkamiausi tvirtai plėvelei pagaminti. Krakmolo kilmės polimerai pasižymi prastesnėmis mechaninėmis savybėmis, tačiau labai greita suirimo trukme. Pasaulyje augant tvarumo aktualumui, ši alternatyva tampa būtina apsvarstyti, siekiant trumpo galiojimo laiko ir lengvo suirimo polimerinės pakuotės. Gaminant palyginamąsias plėveles, geriausiomis mechaninėmis savybėmis pasižymėjo plėvelės sudarytos iš bulvių krakmolo ir sujungtos su sorbitolio plastifikatoriumi. Žirnių skaidulos turėjo teigiamą efektą sustiprinant polimero struktūrą, tačiau tiesioginio plėvelės stiprumo nepadidino. Siekiant plėtoti biologiškai skaidžių plastikų gamybą, reikia ieškoti balanso tarp lengvai skylančių, tvirtumą suteikiančių, atsparumą drėgmei padidinančių medžiagų.

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Introduction

Problem – Oil-based packaging for single-use irreversibly pollutes the environment.

Hypothesis – Biodegradable plastics of plant origin, made from potato and corn starch and pea fibres, are a suitable alternative to petroleum-based plastics.

The purpose of the packaging is to protect the product from scratches, spills, and other environmental factors. When assessing the packaging from the point of view of product protection, it must be strong, flexible, and inexpensive. The tightness of the packaging, mechanical properties, directly affects the quality of the stored product. The most commonly used polymeric material in the manufacture of supermarket bags is polyethylene. It is difficult to compete with this material due to its good mechanical properties and cheap price. However, although the properties of polyethylene allow the production of strong packaging that fully protects the product from external factors, it is made based on oil. Polymers made from petroleum products have a particularly long degradation period. This problem is becoming more acute due to humanity's endless coping with the accumulation of plastic waste in soils and oceans. Various steps are being taken to reduce pollution from plastic products: directives, mandatory standards, a deposit system, reusable packaging, and so on. In this work, an alternative to oil-based packaging is proposed - to use short-lived packaging, which is made of biodegradable materials of plant origin. Such a package would be slightly weaker than traditional packages, but its disintegration time is very short at the required temperature and humidity. Sustainability solutions are becoming relevant not only for legislators but also for consumers themselves. Growing education about ecology, environment, consumerism, reveals the sad reality that the comfortable life of humanity has a negative impact on the future. Waste sorting is the first step towards more sustainable use of packaging by consumers, but some packaging is multi-layered and sometimes impossible to recycle (except incineration). When producing the packaging, the manufacturer is often at a dead end when choosing priorities between price, sustainability, processing method, and strength of mechanical properties. Plasticizing, stabilizing, antioxidant materials are often used to strengthen the polymeric structure, making them unnatural or complex in an environment, particularly difficult to bind. It is important to make every effort to develop new and improve existing materials to extract the best possible mechanical properties from a structure that breaks down as easily as possible, avoiding micro-plastics and other long-lasting legacies.

Aim: To design plant-based biodegradable plastic composites in the use of sustainable packaging.

Tasks:

1. Develop comparable plant-based biodegradable plastic composite films;
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4. Investigate further composting, biodegradation, and recycling.

1. Analytical part

1.1. Relevance of plastic packaging pollution in the world

As the world's cities expand each year and the population grows rapidly, plastic-based packaging is being produced and used in ever-increasing quantities. [1]. According to 2018 data, only ~ 9 % of the plastic produced worldwide is recyclable. According to current consumption projections, in 2050, about 12 tonnes of plastic waste could be disposed of in the environment (soil and water). With plastic production growing at the same rate as today, the plastics industry could account for 20 % of global oil consumption [2]. When assessing plastic consumption, it is important to determine its consumption by period and by sector. In Fig. 1 the volumes of global plastic production from 1950 to 2015 are presented. Thus, over 65 years – plastic production has increased 190.5 times [3].

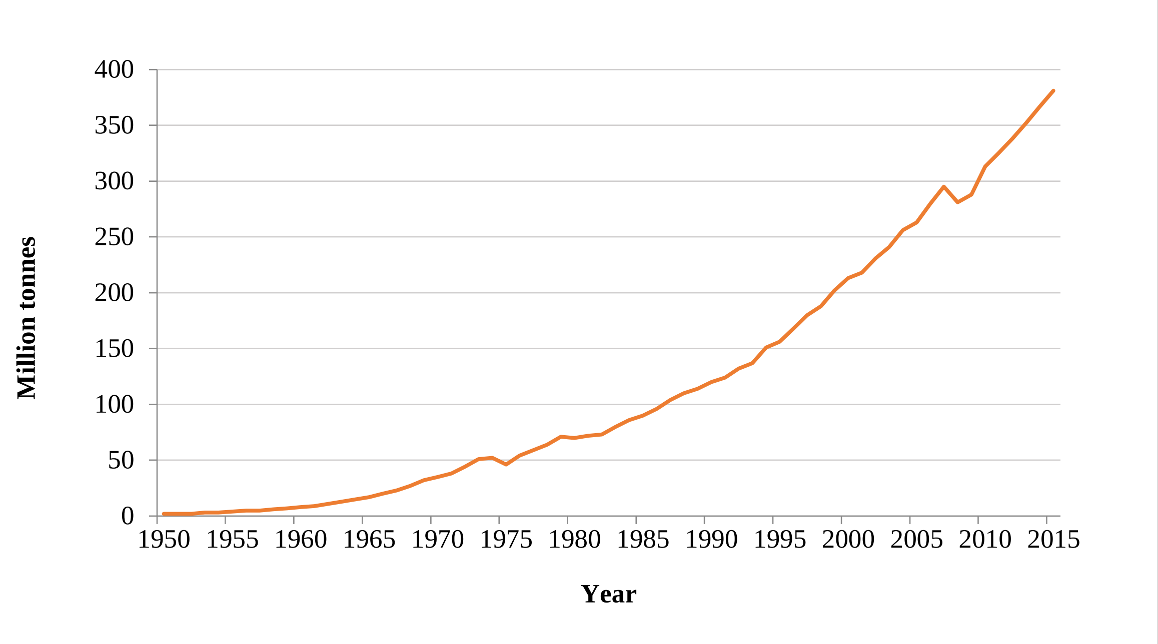

Fig. 1 Global plastic production volumes (1950 – 2015) [3]

In terms of sector-specific production of plastic products in 2015, the packaging industry ranks first, i.e. 146 million tonnes of plastic for packaging was produced in 2015. The building and construction sector ranks second with 65 million tonnes of plastic produced for construction purposes. 59 million tons of plastic were produced for the textile sector. The responsibility of the packaging industry for the highest consumption of plastics in all sectors is a clear sign that past and current policies on the exploitation of plastics are not sufficiently effective. Also contributing to this is the growing birth rate every year, improving economic conditions, rising requirements of the company's quality standards for the customer, so the packaging of the product pays great attention to preserve the product. In some cases, the packaging is an important design accent that affects whether or not the buyer will buy the item. Through the visualization of the packaging, it is possible to express the idea that the company wants to convey, to create a representative presentation. an attractive packaging design can significantly increase purchases compared to a product of the same quality in an unattractive package. Also, for reasons of public hygiene, heavily weighed foods are becoming an increasingly less desirable choice. The packaged product guarantees that it is safer, more hygienic. Sector distribution data are shown in Fig. 2 [3].

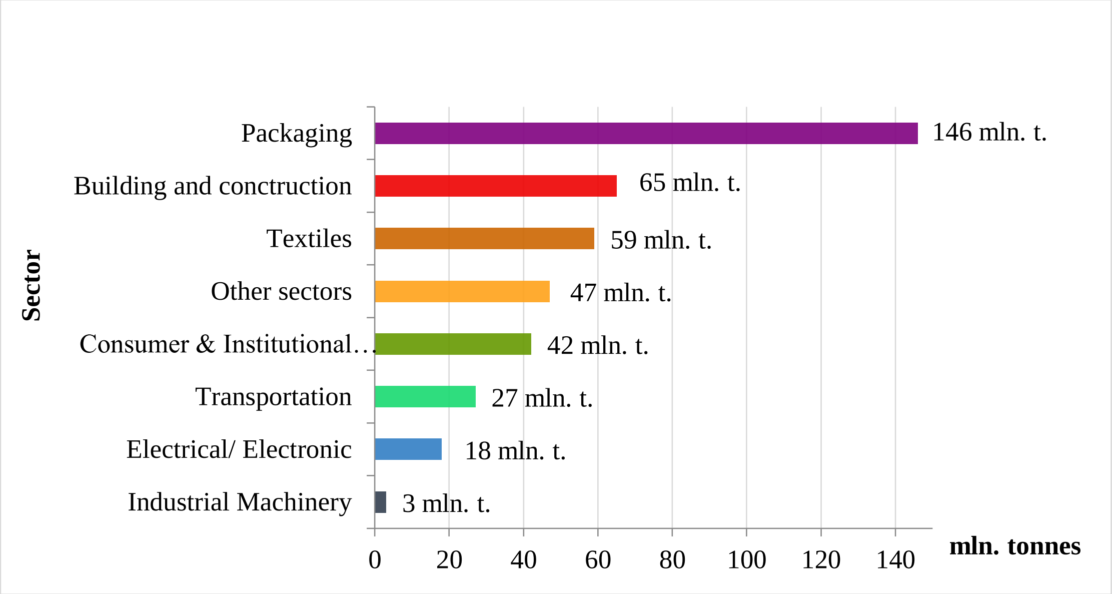

Fig. 2 Primary plastic production by sector in 2015 [3]

Accelerating plastic production and consumption may be affected not only by expanding demographics of increased needs but also by unavoidable causes such as a global pandemic. The global COVID-19 pandemic that began in 2019 has further increased the amount of plastic waste. Disposable sterile plastic products such as disposable gloves, disposable masks, respirators N95, goggles, protective medical suits, masks, hand sterilization bottles, syringes, waterproof aprons, water bottles, takeaway packaging, polyethylene bags, medical test cases are used to protect people from the virus. This increase in medical waste was affected by the promotion of social distance to minimize the chances of getting infected with the virus. Managers of many grocery stores have chosen to wrap fruits and vegetables in plastic film for reasons of consumer hygiene [4]. According to the *World Health Organization*, in 2018 in affluent countries, one hospital bed generates an average of 0.5 kg/day of hazardous medical waste, in poor countries one hospital bed generates an average of 0.2 kg/day of hazardous medical waste [5]. The COVID-19 outbreak is estimated to have caused 1.6 million tons of plastic waste worldwide per day [4]. Plastics are made from petroleum-based products that are made from hydrocarbons. However, often the composition of plastics consists of stabilizers, plasticizers, dyes, which are not environmentally friendly [6]. According to Fig. 2, the packaging sector is the main concern. The main materials used in the manufacture of packaging are shown in Fig. 3 [7].

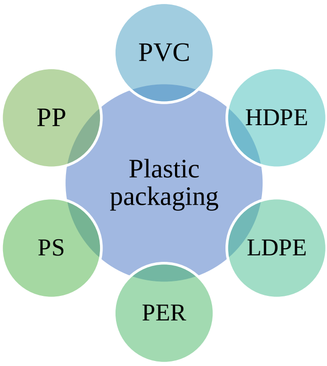

Fig. 3 The most common basic materials for plastic packaging [7]

PVC plastic has one of the strongest mechanical properties. However, due to its composition, it poses serious risks to humans, carcinogenic, mutagenic, genetic alterations, birth defects, and in particular, ecotoxicity. Hazardous substances enter the soil, thus reaching the water. PVC components and their effects are illustrated in Table 1. [8].

Table. 1 Effects of PVC components on humans and the environment [8]

Carcinogenic effects	Inherited mutagenic lesions and malformations	Ecotoxicity
Antimony(III) oxide Cadmium stearate Acetylene black Polychlorinated n-alkanes DEHP Lead carbonate/ stearate Silicon dioxide Talcum powder Chloroethene Sawdust	Stearic acid Various esters of phosphoric acids Various esters of phthalic acids Various adipic acid esters Sebacic acid di-(2-Ethylhexyl) ester Stearamide Dibutyltin dichloride Carbon black pigment Tributyltin chloride Dioctyl phthalate Quaternary ammonium compounds Soybean oil (epoxidized)	Stearic acid Various esters of phosphoric acids Various esters of phthalic acids Triacetin Various adipic acid esters Stearamide Dibutyltin dichloride Tributyltin chloride Soybean oil (epoxidized) Quaternary ammonium compounds Glycerol monostearate

HDPE (High-density polyethylene) pipes have high corrosion resistance, high hydraulic efficiency. [9]. HPDE and LPDE (Low-density polyethylene) plastics are considered safe for food packaging, but the stabilizer nonylphenol used in the production of HPDE reacts with ultraviolet light (such as the sun during the daytime) to break down into substances that can impair human endocrine and hormonal activity. About 29 % of HPDE manages to recycle and remanufacture packaging [10]. PS (Polystyrene) production requires benzene, which is a carcinogenic substance. Styrene formation has a negative impact on male and female reproductive health [11]. Being a carcinogen, styrene damages the brain and nervous system. The risk of styrene leaking into food arises when the food is hot and humid [10]. Given the existing risks of petroleum-based plastics, biodegradable plastics of plant origin are being developed and used as an alternative. In 2019, 1.952 million tonnes of bioplastics (partially degradable) were produced worldwide, of which 1.051 million tonnes were biodegradable (53.8 %). In 2020, 2.111 million tonnes of bioplastics (partially degradable) were produced, of which 1.227 million tonnes were biodegradable (58.1 %). By 2025, the production of biodegradable plastics is projected to reach 1,800 million tonnes. However, bioplastics currently account for ~1 % of all plastics produced per year globally. Biodegradable plastic production forecasts are shown in Fig. 4 [12]. Assessing the possible use of bioplastics, it can be concluded that their use is quite wide. Flexible and rigid packaging, consumer goods, textiles, electronics, agricultural products. However, the most popular and most promising area is the packaging sector. Compared to other products, the packaging is not required to last a long time. For this reason, sensitive structured plant-based polymers can be successfully applied to environmentally friendly packaging while raising the question of whether planetary survival is more important than humanity’s growing needs. Thus, although the primary purpose of packaging is to protect the product, it is important to investigate what the composition of the packaging is made of and what effect it will have on living organisms.

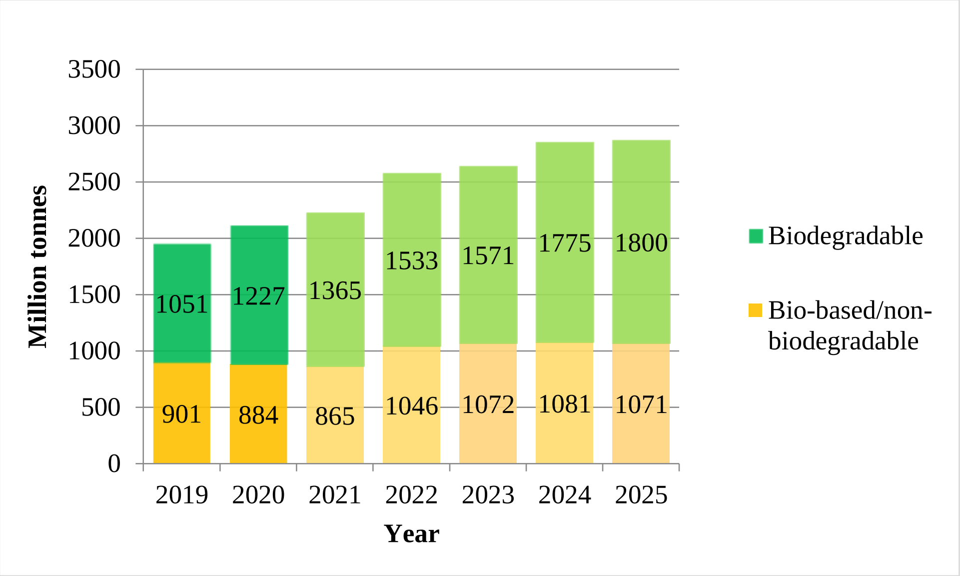

Fig. 4 Global production capacities of bioplastics [12]

Biodegradable plastics derived from biomass benefit from the activity of microorganisms during degradation. The use of the activity of microorganisms in degradation processes can significantly reduce the problem of pollution of plastic packaging when traditional packaging does not decompose on its own for years. At the appropriate temperature, relative humidity, in a suitable environment, biodegradable plastics decompose into biomass, H_2O, and gas. Mechanical properties are important parameters in assessing whether they can compete with petroleum-based plastics. The degradation of biodegradable plastic belongs to the chemical characteristics and functional groups of the polymer, the chemical structure, and the crystallinity of the polymer. Although this type of plastic decomposes easily in the soil and does not pollute it, the degradation process requires certain conditions that are not present in all environments [13]. For biodegradable plastics to successfully adapt to the operational needs of plastics in industry, it is necessary to establish the most important characteristics:

- Standardization of biodegradable plastics (consumer safety);
- Biodegradability specifications;
- Do all open environments/soils have the same conditions for degradability;
- Do microplastic particles has an impact on environmental microorganisms;
- Any factors if biodegradable plastic could have a negative impact on the environment.

1.2. The scientific novelty of the research

One of the most popular materials for developing new environmentally friendly materials and researching their properties is starch. In the development of starch-based plastics, starch is treated with plasticizers and an extrusion process is used. Sometimes a biodegradable mixture is used alone or in combination with petroleum-based polymers. In this case, the plastic decomposes harder but has better mechanical properties. Starch-based plastics are used in agro films, organic waste for compostable bags, and also used in convenience store bags. End-of-life composting and anaerobic digestion of starch-based plastics is the right way to solve the problem of plastic accumulation. There is a possibility to produce biogas, to perform composting processes [14]. The substance is recognized as biodegradable when it breaks into CO_2, H_2O, CH_4 molecules, and biomass at the end of the biodegradation. Plastic bags or packaging in supermarkets are often referred to as ‘degradable’, ‘oxo-degradable’ or ‘biodegradable’. However, the biodegradable material should be one that does not contain non-biodegradable additives (such substances are usually found in stabilizers, dyes, and plasticizers) [15]. Recently, with consumer interest in more sustainable packaging solutions, there has been a lot of misunderstanding about terminology. *Biodegradable plastic* is a polymer that can degrade under natural conditions by living microorganisms from the same material it is made of. *Bioplastics* are described as made from renewable natural raw materials, but they are usually not easily composted, even though they are made from natural materials. *Compostable plastic* requires special environmental conditions (certain temperature, relative humidity, soil acidity, etc.) to become completely degraded. Thus, while biodegradable plastic can degrade in the natural environment, compostable plastic needs special conditions [16]. The most common types of biodegradable plastics are shown in Fig. 5 [17].

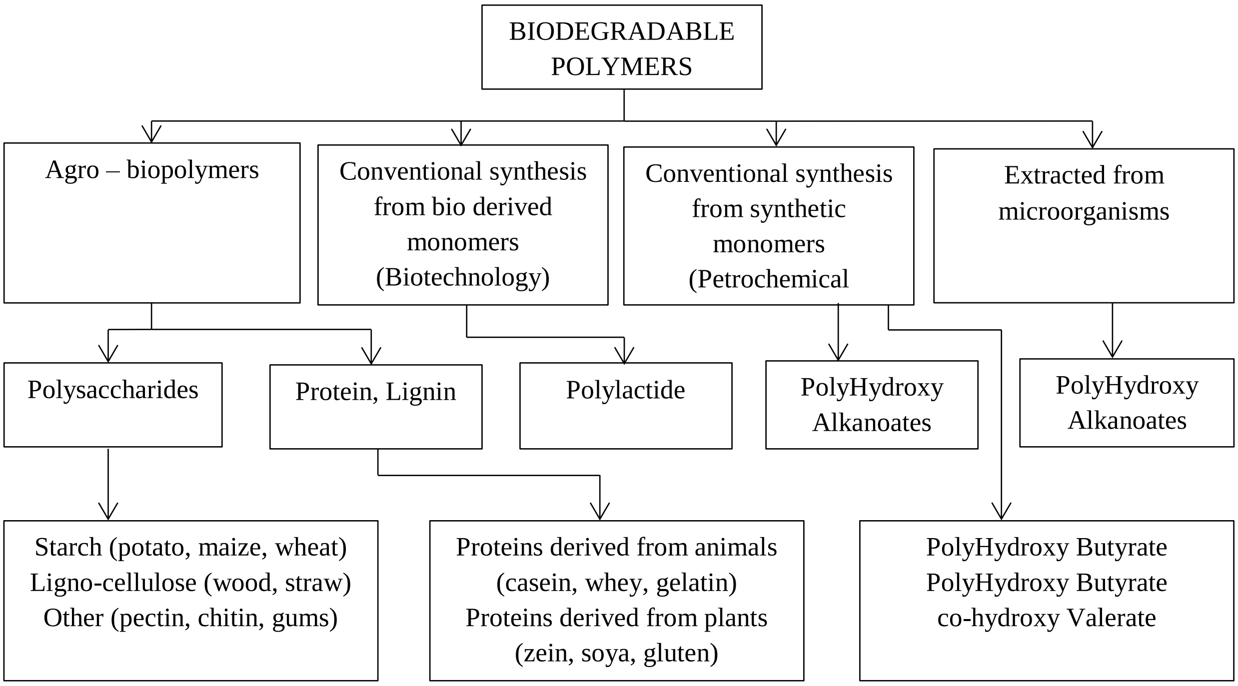

Fig. 5 Classification of biodegradable polymers [17]

Starch can be found in nature. They are produced in the leaves of plants in the daytime and accumulate when it is dark. Starch is consisting of two types of glucose polymers - amylopectin and amylose (Fig. 6). The appropriate ratio of amylopectin to amylose has a significant effect on the physical and chemical properties of starch or starch-based products, which further affects its functionality and potential use. Higher amylose content contributes to the film's strength. Branched amylopectin results in lower stress / mechanical strength film properties that can be improved by the use of plasticizers such as sorbitol or glycerol. Starch-based polymers have higher bioavailability and oxygen barrier properties. The starch of natural origin predominates as a heteropolymer in nature and is used in various forms in admixture with conventional polymers to produce a biological polymer. Starch is a white powder that does not have a specific smell and taste. therefore, the packaging product does not acquire an undesirable odour and colour. starch-based plastics can be dyed with natural dyes, but this requires colour-fixing agents so that the product does not change colour when oxidized [17].

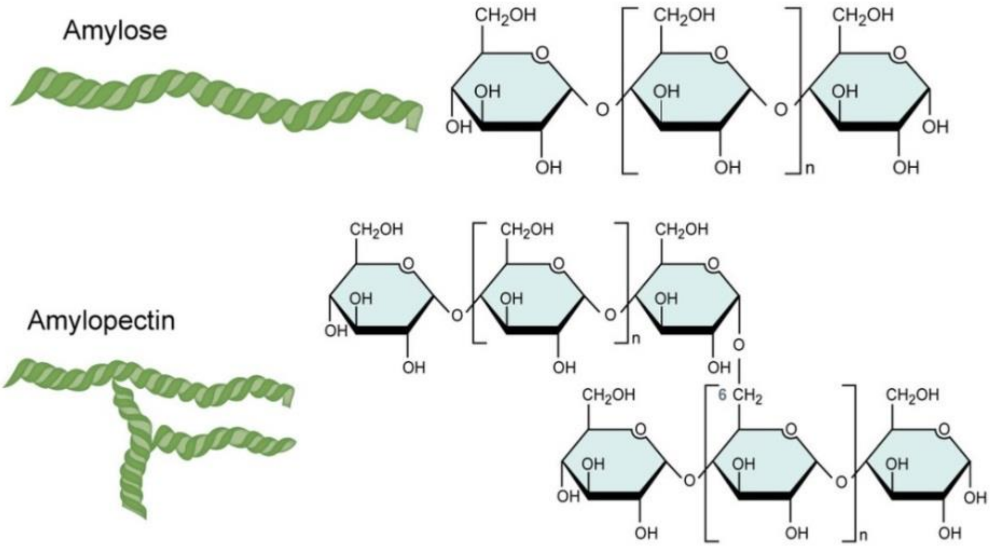

Fig. 6 Composition of starch [18]

Starch-based plastics can decompose from 14.2 % to 85 % under anaerobic conditions, at 23-58 ° C, in 72-236 days, depending on the environment (soil, wet sand deposits, special compost). Different types of cellulose-based plastics can decompose from 35 % to > 80 % in 14 to 154 days (depending on the decomposition environment - in a mixture of municipal solid waste or compost adapted for this purpose. PLA-based plastics decompose from 10 % to 100 %, in 28 to 98 days (depending on composting type) [17]. Based on those results, it was decided to continue to focus on starch-based polymers in this work, because of plant origin. Biodegradable polymers are characterized by the presence of oxygen and nitrogen in the polymer, so the polymer structure is easily degradable. Challenges for the commercialization of starch-based packaging include:

- Overcoming mixing problems with high starch content;
- Avoidance of deterioration of mechanical properties in case of high starch content
- Reduction of costs when starch polymer blends consist of a low starch content [19].

1.3. Analysis of similar research

Shafqat, N. Al-Zaqri, A. Tahir, A. Alsalme *(2021)* during a scientific experiment investigated the use of natural agricultural materials for the production of biodegradable plastics. The comparative samples were of two types: *banana peel starch* samples and *composite* samples (consisting of banana peel, corn, and rice starches). The experiment aimed to investigate the use of readily available, abundant, biodegradable waste as a filler in polymer films, thus enhancing the mechanical properties of plastics. The materials used in addition to starch were: ethanol, acetic acid (glacial), distilled water. As a plasticizer choose glycerol and sorbitol. Tree dust and potato peel powder were used as fillers [20]. Twelve samples of banana peel starch and 12 samples of composite composition (banana peel, corn, and rice starch) were prepared. Different amounts of plasticizers and fillers are used. Acetic acid content (5 %) used in all formulations. No fillers or plasticizers were used for the control. Moisture content, water absorption, water and alcohol solubility, biodegradation test were studied. Mechanical behaviour results of *banana peel* starch samples are shown in Table 3, the results of the *composite* samples are shown in Table 2.

Moisture content

When evaluating mechanical properties of vegetable starch-based polymers, it was observed that both BPS (37,63 %) and $BRC_{comp.}$ (35,52 %) starches combined with sorbitol provided the highest moisture content compared to other compositions. This can be explained by the fact that glycerol is formed by a hydroxyl group that has similarity to water molecules [21], this way it is possible to create hydrogen bonds and attach water to the structure. Thus, as the amount of sorbitol as a plasticizer increases, so does the amount of moisture. Samples with starch and fillers had the lowest moisture content – BPP (1,46 %), $BRC_{comp.}$ (4,65 %). Moisture in the polymer is required for elasticity, non-drying properties.

Water absorption

In assessing water absorption, both BPP (218,39 %) and $BRC_{comp.}$ (215,79 %), the highest value of absorption was observed in the starch samples (control). More starch a film contains, more it absorbs water. Because of the hydroxyl groups, starch can gelatinize, breaking down the granules, allowing water to diffuse [22]. This means that this type of polymer has low moisture resistance and could be more difficult to adapt to the packaging sector. Samples consisting of starch and sorbitol - BPP (85,66 %), $BRC_{comp.}$ (87,9 %) – absorbed the least amount of water. It was influenced by the hydroxyl group of sorbitol. More sorbitol it contains, lower the absorption is.

Solubility in water

The highest results of water solubility were observed in samples consisting of starch and glycerol – BPP (85,57 %), $BRC_{comp.}$ (67,65 %). This is because crystalline starch molecules consist of hydrogen bonds, therefore, starch granules are usually insoluble in cold water [23]. The lowest values of water solubility were found in samples from starch and filler - BPP (25,35 %), $BRC_{comp.}$ (8,97 %). Because the potato peel powder and wood dust) are not soluble in water, also the potato peel powder contains starch, so the starch granules do not dissolve at normal room temperature. Wood dust is made up of cellulose, which is insoluble in water [24].

Solubility in alcohol

The highest values of alcohol solubility were found when starch was combined with plasticizers - BPP (74,44 %), $BRC_{comp.}$ (60 %). At normal temperatures, starch is insoluble in alcohol and sorbitol is slightly more soluble in alcohol than water. Samples from starch and fillers had the lowest values – BPP (23,46 %), $BRC_{comp.}$ (8,05 %). This is because the fillers (wood dust and banana peel powder) are made up of cellulose and starch, which are insoluble in alcohol.

Biodegradation

Polymer degradation was determined by molecular weight, structure, and other physicochemical properties. The highest biodegradation rates were found in samples consisting of starch and glycerol – BPP (81,41 %), $BRC_{comp.}$ (79,09 %). This can be explained by the fact that the addition of a plasticizer increases water absorption. The lowest biodegradation results were found in samples consisting only of starch (control) - BPP (12,07 %) and starch with fillers $BRC_{comp.}$ (31,94 %). As the amount of plasticizer increased, the degree of biodegradation increased. As the amount of fillers increased, the degree of biodegradation decreased.

Table. 2 Properties of BPP and BRC_{comp} films [20]

BPP (banana peel starch)									
Amount of H_2O, %		**Absorption of H_2O, %**		**Solubility in H_2O, %**		**Solubility in Alcohol, %**		**Biodegradation, %**	
Min	*Max*	*Min*	*Max*	*Min*	*Max*	*Min*	*Max*	*Min*	*Max*
Starch, filler (1,46)	Starch, glycerol (37,63)	Starch, sorbitol (85,66)	Starch (218,39)	Starch, filler (25,35)	Starch, glycerol (85,57)	Starch, filler (23,46)	Starch, glycerol, sorbitol (74,44)	Starch (12,07)	Starch, glycerol (81,41)
BRC_{comp} (banana peel, rise, corn starches)									
Starch, filler (4,65)	Starch, glycerol (35,52)	Starch, sorbitol (87,9)	Starch (215,79)	Starch, filler (8,97)	Starch, glycerol (67,65)	Starch, filler (8,05)	Starch, glycerol (60)	Starch, filler (31,94)	Starch, glycerol (79,09)

An analysis of the data from scientists suggested that both sorbitol and glycerin were suitable plasticizers for biodegradable starch polymers. Sorbitol showed a positive effect on the moisture content in the sample (the more sorbitol, the higher the humidity), and sorbitol also influenced the resistance to water absorption. Glycerin gave the samples a higher water solubility, but a positive effect on biodegradation. Both plasticizers have in samples to increase the solubility in alcohol. The fillers in the polymer samples gave the lowest moisture values, and such samples were the most soluble in water and alcohol, and hardly biodegradable. Based on these results, sorbitol and glycerin will be selected as plasticizers in the following sections.

1.4. Engineering methods that are used for solving similar scientific/engineering problems

1.4.1. Solutions that are used in the market

Mater-bi™

Novamont, a company founded in Italy in 1989 with subsidiaries (*Biobag International AS*, *Mater-Biotech SPA*, *Tecnogen S.p.A.*, *Mater-Biopolymer Srl.*), was one of the first to make the most extensive use of its biodegradable thermoplastic Mater-bi™. This brand consists of patented technologies from starch, cellulose, and vegetable oil. This plastic is used to make bags for wholesale and retail chains, the food packaging sector, personal care, the agricultural sector [25]. *Biobag International AS* an example of the bag produced is shown in Fig. 7. While the exact composition of Mater-Bi® remains a trade secret, some indications are that the main ingredients are corn starch and various synthetic blends, including plasticizers and biodegradable hydrophilic materials made from synthetic polymers. However, there are more types of Mater-Bi® from other ingredients [26]:

- *(Y type)* the products may consist of a mixture of starch and cellulose acetate with properties similar to polystyrene;
- *(A type)* consisting of copolymers of thermoplastic starch and polyvinyl alcohol;
- *(V type)* consists of > 85 % thermoplastic starch with high water solubility;
- *(Z type)* consisting of a matrix of biodegradable polyester polycaprolactone;
- *(N type)* consists of a polymer matrix – Polybutylene adipate terephthalate.

Fig. 7 Bag made of *Mater-bi*™

BIOPLAST

The company established in Germany *BIOTEC GmbH & Co. KG* has been developing, researching, and producing sustainable raw materials for packaging manufacturers since 1992 – biodegradable plastic resin made from renewable plant resources. Areas of application include supermarket and garbage bags, pharmaceutical encapsulation products, and other packaging solutions. Plasticizers are not used in the production process (except *BIOPLAST TPS*®). The company's goal is to produce polymer products with the largest possible amount of materials of biological origin [27]:

- *BIOPLAST GF 106/02* type seeks 23 %;
- *BIOPLAST 500* tipas type 50 %;
- *BIOPLAST GS 2189* type 69 %;
- BIOPLAST TPS® 100 %.

The company supplies a wide range of biodegradable raw materials for plastic production internationally. Latest products of the last 6 years:

- *BIOPLAST FRCL 500 (created in 2019)* – the material is suitable for the production of a transparent inflatable film. Can be composted at home. < 50 % of biological origin;
- *BIOPLAST 105 (created in 2018)* – suitable for hot molding type packaging and 3D printing. Suitable for sheet production and blown film extrusion methods (Fig. 8). Thermoplastic and translucent material consists of 67 % of biological origin. Easily decomposes in industrial composting environments;
- *BIOPLAST FRCL* 400 *(created in 2017)* – the material is suitable for the production of a transparent inflatable film. Can be composted at home. < 40 % of materials of biological origin;

- *BIOPLAST 300 (created in 2016)* – this thermoplastic material is adaptable for the production of a transparent inflatable film, for the production of very thin films (~ 10 µm), for bags. Can be composted at home. Complies with *EN 13432* and is fully compostable and biodegradable. The product contains < 30 % materials of biological origin. Plasticizers are not used in this material and include natural potato starch and other polymers of biological origin;
- *BIOPLAST 400 (created in 2015)* – the material is suitable for the production of a transparent inflatable film. Suitable for the production of very thin films (~10 µm), for bags. Can be composted at home. Complies with *EN 13432* and is fully compostable and biodegradable. The product contains < 40 % biological materials. Composed of natural potato starch and other polymers of biological origin [27].

Fig. 8 Example of *BIOPLAST 105* end product – packaging [27]

BIOTEC the company's products are based on the *EN 13432* standard, these products biodegrade in less than 180 days. Industrial compost decomposes in less than 90 days. According to ASTM D6866, the main material of BIOPLAST products is plant materials, mainly potato starch [27].

Ecoflex®

This material is manufactured by the company BASF. These are completely biodegradable polyphenylene oxides in a mixture containing starch or polylactic acid. This biodegradable material has been designed to perform similar functions to polyethylene - flexible, strong, tear-resistant, moisture-resistant. This raw material is intended for short-term packaging such as organic waste bags, food film, cups. The aim is to improve the barrier properties so that the film "breathes" and is suitable for packaging fresh vegetables and fruits. This is a fossil-based, man-made polyester but yet is completely biodegradable due to its chemical structure [28].

1.4.2. Researchers conducted studies with biodegradable bags

J. B. Olivato, M. V. E. Grossmann, A. P. Bilck, F. Yamashita In an experiment conducted by researchers, mixtures of cassava starch and polyester were investigated to explore the potential for bag production. To evaluate tensile strength, puncture resistance, tensile elongation, and seal strength of cassava and PBAT blend films using tartaric acid as a stabilizer. Materials used: natural cassava starch, polyester PBAT Ecoflex®, glycerol (plasticizer), tartaric acid (mixture stabilizer). Five different compositions were made with the same materials but in different proportions. The influence of plasticizer and stabilizer on the composition was evaluated, the total mixture ratio of starch and PBAT was used 55:45. The production of films starts with the granulation of the mixture of selected materials. The selected bead diameter is 2 mm. A laboratory single-screw extruder was used to produce the pellets [29]. The schematic granulation process is shown in Fig. 9.

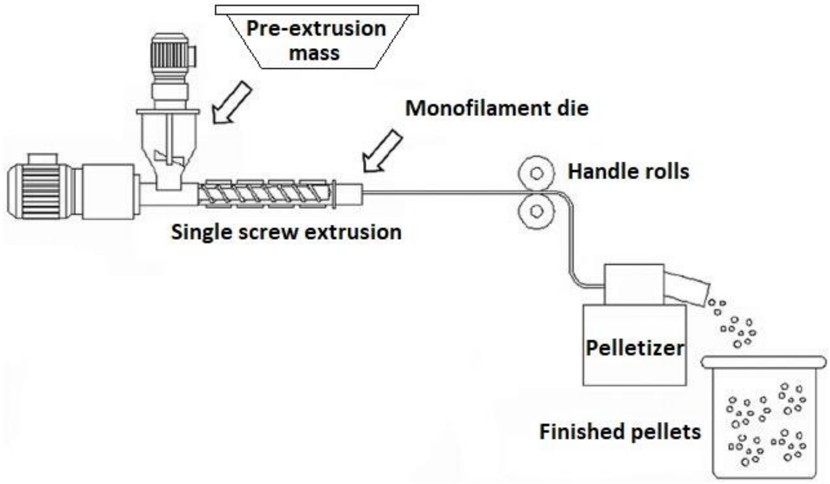

Fig. 9 Example of single-screw extruder [30]

After the first extrusion, the process was repeated a second time to obtain the blown films. The film thickness was adjustable from 100 to 150 µm. For each of the five compositions, ten specimens measuring 25x80 mm were prepared. A schematic process of film blowing is shown in Fig. 10.

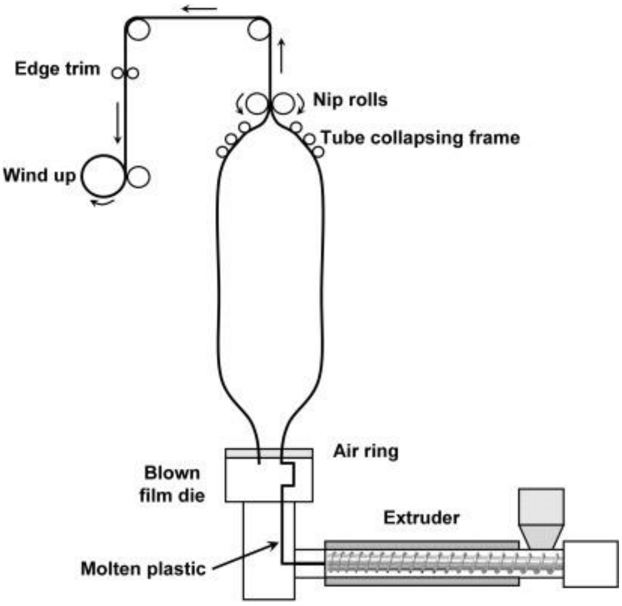

Fig. 10 Example of film blowing process [31]

Sealing strength

Sealing the edges of the film is a very important feature to produce a quality bag or other packaging because the product must retain the product, prevent it from leaking, falling out. The minimum value of the tightness strength results was reached at 190 N and the maximum value at 720 N. Higher values of tightness force were observed in those samples with the lowest tartaric acid concentration, depending on the proportions of the compositions used. No cyclic effects of glycerol with increasing or decreasing concentrations were observed [29].

Puncture force

This characteristic is relevant for packages that have edges, tips, and other protruding edges that can self-pierce the package. The minimum value for the puncture force is 10.99 N for higher glycerol and the maximum is 24.36 N for higher tartaric acid. Thus, the plasticizer reduces the strength of the puncture force by reducing the intermolecular forces between the polymer chains [29].

Elongation

The minimum elongation value was 176.92 % and the maximum 736.68 %. Samples with higher glycerol content had higher molecular activity, thus this resulted in a higher elongation value [29].

Moisture content

No direct effect on moisture was observed during the monitoring of the films without plasticizers. The films plasticized with glycerol increased moisture content due to hydrophilicity. Glycerol creates active sites in the film matrix where hydroxy groups of glycerol are observed where adsorption of the water molecule is possible [29].

Flexibility

The addition of glycerol increased the flexibility of the film, reducing tear forces and increasing the film deformation. As previously described, glycerol interference in the polymer chain association facilitates their slip. Thus, the addition of plasticizer modified the mechanical profile of the film into a flexible material, as the tensile strength decreased and the elongation at break increased accordingly ($p < 0.05$). Acetylated starch formed more resistant matrices compared to tonic starch films [29].

Tensile strength

The tensile strength of films containing acetylated starch was significantly ($p < 0.05$) higher than that of native starch films, regardless of glycerol addition. Although acetyl groups interfere with chain alignment, they resulted in a more resistant matrix as the hydrogen bond between the aligned polymer chains was enhanced [29].

Young's modulus

Young's modulus showed a similar trend in tensile strength. Glycerol supplementation improved mechanical behaviour by increasing elongation at break, although this increase was significant ($p <0.05$) only for acetylated starch films [29].

As the tartaric acid concentration increased, the sealing strength of the edges decreased. Increasing the glycerol content results in lower puncture force values, and increasing the tartaric acid content results in higher puncture force values. Glycerol had a positive effect on elongation values. Films plasticized with glycerol increased the moisture content. The addition of glycerol increased the flexibility of the film, reducing tear forces and increasing film deformation. Acetylated starch formed more resistant matrices compared to tonic starch films. The tensile strength of films containing acetylated starch was significantly higher. Glycerol had a positive effect in increasing elongation during fracture.

2. Experimental part

The experimental part aims to create a prototype of the films discussed in the articles with selected materials at the laboratory level. The samples are then compared with each other to determine which composition has the most potential to achieve production.

The aim:

- To create comparable biodegradable plant-based plastic films;
- To measure the thickness of the films;
- To compare the tensile strength;
- To compare Young's module;
- To compare the contact angle;
- Using a microscope, evaluate the distribution of oxygen in the film.

2.1. Materials used

Materials used for the film molding: potato starch, maize starch, pea fibres (extracted from the kernel), sorbitol, glycerol, vinegar (9 %), distilled water.

Potato starch

Potato starch is produced by crushing potatoes until leucoplastic grains are released. The starch is washed and dried. The resulting powder is the final product. The heated starch paste is completely clear, odourless, easily soluble in water. The production process is called biorefining. Compared to other starches, potato starch has the highest viscosity, the lowest gelatinization temperature, and the maximum granule size. Potato amylose has the highest molecular weight values and the lowest level of branching. Potato starch varieties with a high starch content are most often selected for starch extraction. Polarizing microscopic view of starch granules is shown in Fig. 11 [32].

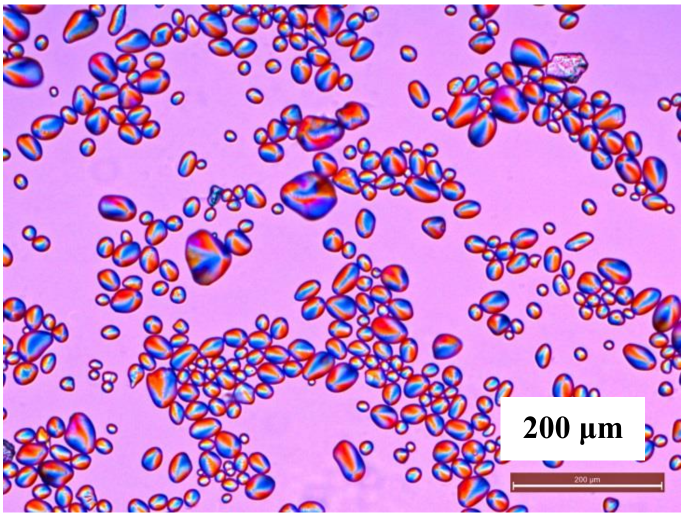

Fig. 11 Potato starch granules [32]

The main composition of the potato is water. Potato starch is mainly used in the food industry and as a functional substance in plastic products [32]. The most common nutritional composition found in potatoes is shown in Table 3.

Table. 3 Chemical composition of potato [32]

Composition	Value, %
H_2O	72
Potato starch	22
Protein	2
Fibre	1
Sugars	1
Various nitrates and amino acids	1
Organic acids	1

Already produced modified potato starch *Alojas* was used (produced by *Aloja Starkelsen SIA*). Potato starch specifications are provided in Table 4.

Table. 4 Parameters of potato starch *Alojas* [33]

Parameter	Value
Moisture content	18.0 – 20.5 %
pH value	4.5 – 7.0 (approx 6.0)
Solubility	Completely soluble after boiling
Brabender viscosity, at 95°C	200 – 500 BU
Brabender viscosity, after 20 minutes at 95°C	300 – 700 BU
Brabender viscosity, at 30°C	700 – 16000 BU
Acetyl value (titration)	Max. 2.5 %

Maize starch

Of maize grains endosperm already produced modified waxy maize starch *CLEARAM® CH 40 20* (acetylated di-starch adipate) was used (produced by *Roquette Frères)*. To make cornstarch, the corn is soaked for 1-2 days. Gentle fermentation takes place. While still soaking, the germ in the corn is separated from the endosperm and they are ground one by one. The starch is separated from the remaining materials (germ, fibre, gluten) and begins to dry. Maize starch chemical parameters are provided in Table 5 [34].

Table. 5 Chemical composition of maize [34]

Composition	Value, %
H_2O	16,7
Protein	9,91
Fat	4,45
Ash	1,42
Carbohydrate	69.60-79.83
Fibre	2,66

Already produced maize starch (*CLEARAM® CH 40 20*) physical parameters are shown in Table 6 [35].

Table. 6 Parameters of maize starch *CLEARAM® CH 40 20* [35]

Parameter	Value
Loss on drying	13 % max.
pH value	4.5 – 6.5
Brabender viscosity	
- Peak	470 to 570 BU
- Drop	-60 to -120 BU
Acetyl value (on DS)	1.5 – 2.1 %

Potato and maize starches have certain properties. During the production of the polymer from starch, water osmosis, gelatinization, melting, crystallization take place. Gelatinization processes involve two stages [36]:

- An unchanged starch molecule that can still return to its original state combines with hydrogen (under the action of Van del Wals) through hydrogen bonds. This stage starts at 20 °C and lasts until it reaches 60 °C;
- The hydrogen bond is broken and the water molecule attaches itself to the hydroxyl bond of the starch molecule, the crystals dissolve and the starch granules decay. This stage starts at 60 ° C and lasts until it reaches 90 °C [36].

Pea fibres

Trade name of yellow pea kernel fibres – *Emfibre EF 200*, produced by *Emsland Group*. Pea fibres were used as a filler to enhance the strength of the film. Parameters of pea fibres are shown in Table 7.

Table. 7 Parameters of pea fibres *Emfibre EF 200* [37]

Parameter	Value
Moisture	10 – 12 % (approx. 10)
pH value	4.0 – 7.0 (approx. 5)
Particle size (< 100 μm)	Max. 20 %

Sorbitol

NEOSORB® 70/70 B liquid sorbitol at 50 % concentration was used (produced by *Roquette Frères)*. Sorbitol is an aqueous solution of polyols at 70 % total solids, prepared by catalytic hydrogenation of starch hydrolysate. Sorbitol in films acts as a plasticizer increases moisture, plasticity, and elasticity, prevents drying, crumbling, provides softness. Sorbitol parameters are shown in Table 8 [38].

Table. 8 Parameters of sorbitol *NEOSORB® 70/70 B* [38]

Parameter	Value
Dry substance	69.5 – 70.5 %
Water content	29.5 – 30.5 %
Sorbitol	50 % min
Reducing sugars	0.15 % max.
pH in solution	5 – 7

Glycerol

Liquid glycerol (~99,5 %) was produced by *UAB „Berchem“*. Glycerol in films acts as a plasticizer increases moisture, plasticity, and elasticity, prevents drying and crumbling, provides softness. Glycerol parameters are shown in Table 9 [39].

Table. 9 Parameters of glycerol [39]

Parameter	Value
Content of basic material, %	>99,8
Fraction of water mass, %	<0,15

2.2. Preparation of composite films

Comparative films were prepared from potato and corn starches, different plasticizers (sorbitol or glycerol), and yellow pea fibres, whether or not mixed together. A planned scheme was followed in the production of comparable films is shown in Fig. 12.

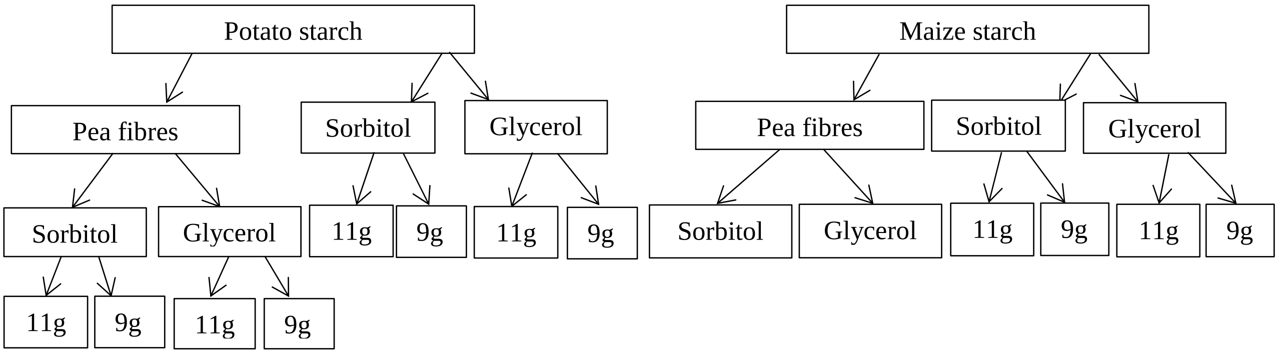

Fig. 12 Scheme for the development of comparative films

Thick polymer mass was transferred to a glass sheet and mechanically distributed manually with a wide spatula. The manual method was used to extract the largest possible useful area without disturbing the polymer structure, which could behave sensitively during the experiment. Drying was performed for 48 hours at 21-22 ° C with a relative humidity of 45-55 %. The dried film began to tear off the sheet when holding the edges. For the accuracy of the results, five samples were prepared for each composition. The compositions of the films produced are given in Table 10.

Table. 10 Composition of comparable plant-based biodegradable plastic films

No. 1	***No. 2***	***No. 3***	***No. 4***
H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Pea fibres 1,2 g Sorbitol 11 g	H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Pea fibres 1,2 g Glycerol 11 g	H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Sorbitol 11 g	H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Glycerol 11 g
No. 5	***No. 6***	***No. 7***	***No. 8***
H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Pea fibres 1,2 g Sorbitol 11 g	H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Pea fibres 1,2 g Glycerol 11 g	H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Sorbitol 11 g	H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Glycerol 11 g
No. 9	***No. 10***	***No. 11***	***No. 12***
H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Pea fibres 1,2 g Sorbitol 9 g	H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Pea fibres 1,2 g Glycerol 9 g	H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Sorbitol 9 g	H_2O 60 ml Vinegar (9 %) 6 ml Potato starch 9 g Glycerol 9 g
No. 13	***No. 14***	***No. 15***	***No. 16***
H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Pea fibres 1,2 g Sorbitol 9 g	H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Pea fibres 1,2 g Glycerol 9 g	H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Sorbitol 9 g	H_2O 60 ml Vinegar (9 %) 6 ml Maize starch 9 g Glycerol 9 g

2.3. Methodology

Thickness measurement

Thickness was measured with a *Lonis Schopper Leipzig* device with an accuracy of ±0,01 mm. The sample was spread between two plates of the device and was pressed with a constant force. The arrow on the meter indicated the distance in millimetres between the detached plates. The average of 3 different film location measurements was derived. The micrometre device is shown in Fig. 13.

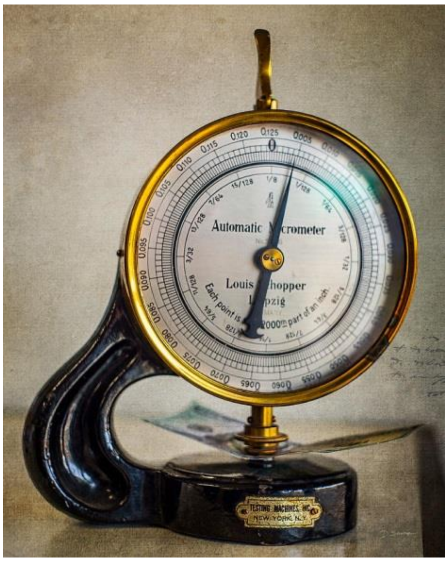

Fig. 13 Micrometer device [40]

Maximum force

The maximum force is calculated from the stress *(N)* and strain (%) diagram results. The load (which created load stress on the specimens) was increased until the samples reached their maximum values. Both ends of the specimens were compressed with the device clamps. For maximum force test *Zwick Z005* (A.S.T. GmbH Dresden) material testing machine were used (Fig. 14). Which parameters were 2.0 mV/V = 200 N. For the experiment, 5 pieces of each type of film were taken, however, *No. 12* were – 4 samples, and *No.* 15 – 3 samples due to the hard rip-off process. Dimensions of one sample: 19 cm x 2 cm.

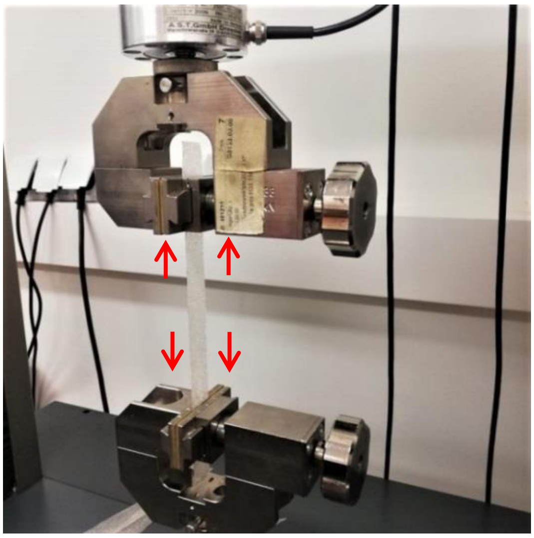

Fig. 14 Zwick Z005 (A.S.T. GmbH Dresden) tensile testing machine

Maximum force test is very important for sensitive, thin and fragile materials. In this way, it can be decided whether the test sample can be strong enough to meet the requirements for mechanical properties of the packaging. Maximum force – FH (*N*) calculated according to the (1) formula.

$$FH = \frac{force\ (N)}{area(m^2)} \qquad (1)$$

Breaking force FB *(N),* calculated according to the (2) formula:

$$FB = \frac{tensile\ extension\ (m)}{lenght\ of\ the\ sample\ (m)} \qquad (2)$$

Young's modulus

Determines the compressive or tensile strength of a material. It is defined as the ratio of stress to relative elongation (deformation). The specimen deforms elastically when a large load is applied when it is stretched. The elastic deformation is reversible (after removal of the load, the material returns to its original shape). At almost zero stress and strain, the stress-strain curve is linear. The higher the modulus, the more tension is required to create the same amount of tension; an idealized rigid body would have an infinite Young modulus. Conversely, a very soft material, such as a liquid, would deform without force and would have zero Young's modulus. Young's modulus – E_{mod} (*GPa*) calculated according to the (3) formula:

$$E_{mod} = \frac{\Delta stress}{\Delta strain} \qquad (3)$$

Contact angle with water

The smaller the contact angle, the more hydrophilic the substance is considered (water accepting). A liquid wets the surface of a solid when its surface tension is less than the surface tension of the solid. The measurement was conducted based on (*Karahan's, 2007*) BV S1008 method. Values of contact angle are shown in Table 11.

Table. 11 Polymer film surface treatment level according to *ASTM D5946* standard

Level of surface finishing	Contact surface angle, °
No processing or slight processing	>90
Small	85 – 90
Medium	78 – 84
High	71 – 77
Very high	<71

Evaluation of reliability of results

The standard deviation measures the scatter of the data set relative to its mean. The standard deviation calculated as the square root of the variance by determining the variation of each data point to the mean. If the data points are further away from the mean, there is a larger deviation in the data set [41]. The standard deviation calculated according to the (4) formula.

$$\sigma = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_i - \mu)^2} \qquad (4)$$

σ – the standard deviation;
μ – the mean of all the values in the population;
n – the number of data points;
x_i – each of the values of the data.

2.4. Results of experimental research

The results of the study included visual observation of the films produced, maximum force values, Young’s modulus, measurement of contact angle with water, thickness measurement, and microscopic observation to find air gaps. The results are presented in the following sections.

2.4.1. Visual assessment

The visual evaluation was performed after the samples were dry. A view of the samples is provided in Fig. 15. Descriptions are provided in Table 12. Of the 16 films, 9 suitable for further testing were obtained, i.e. 56.25 % positive samples were obtained. Almost all samples from corn starch were unformed. This may have been influenced by the choice of the wrong temperature or the wrong plasticizer to plasticize the corn starch, as the films appeared visually crumbled. Films with pea fibres had a rougher, grainier surface. It can therefore be concluded that a smaller fraction of pea fibres would be more suitable. Samples without pea fibres showed complete transparency. Films with a stronger structure during tearing provided sufficient useful area to obtain 5 specimens. However, the weaker films broke during the tearing or cracks had formed in some places already during drying. Therefore, less than 5 samples were obtained.

Table. 12 Visual evaluation of samples

Sample	Visual description
No. 1	Strong as cartridge paper, rough surface (due to the pea fibres), easily torn off the sheet of glass
No. 2	Wet, rough surface (due to the pea fibres), easily adheres to its edges, sticky
No. 3	Strong as cartridge paper, even, transparent
No. 4	Sticky, 48h drying was not enough, sensitive, soft as vellum paper. The wrinkles visible in the film were formed by the manual casting method when the too thick polymer mass was mechanically spreaded on the glass sheet
No. 5	Did not form a film
No. 6	Did not form a film
No. 7	Did not form a film
No. 8	Did not form a film
No. 9	Strong as cartridge paper, rough surface (due to the pea fibres), easily torn off the sheet of glass. The wrinkles visible in the film were formed by the manual casting method when the too thick polymer mass was mechanically spreaded on the glass sheet
No. 10	Smooth, rough surface (due to the pea fibres), easily tears from the glass sheet, wet, sticky, soft as vellum paper. The wrinkles visible in the film were formed by the manual casting method when the too thick polymer mass was mechanically spreaded on the glass sheet
No. 11	Strong as cartridge paper, easily torn off the sheet of glass. The wrinkles visible in the film were formed by the manual casting method when the too thick polymer mass was mechanically spreaded on the glass sheet
No. 12	48 hours was not enough to dry. After 72 hours completely dried, clear, even. The wrinkles visible in the film were formed by the manual casting method when the too thick polymer mass was mechanically spreaded on the glass sheet
No. 13	Did not form a film
No. 14	Did not form a film
No. 15	Soft as very thin paper, hard to peel from the glass sheets. The wrinkles visible in the film were formed by the manual casting method when the too thick polymer mass was mechanically spreaded on the glass sheet
No. 16	Did not form a film

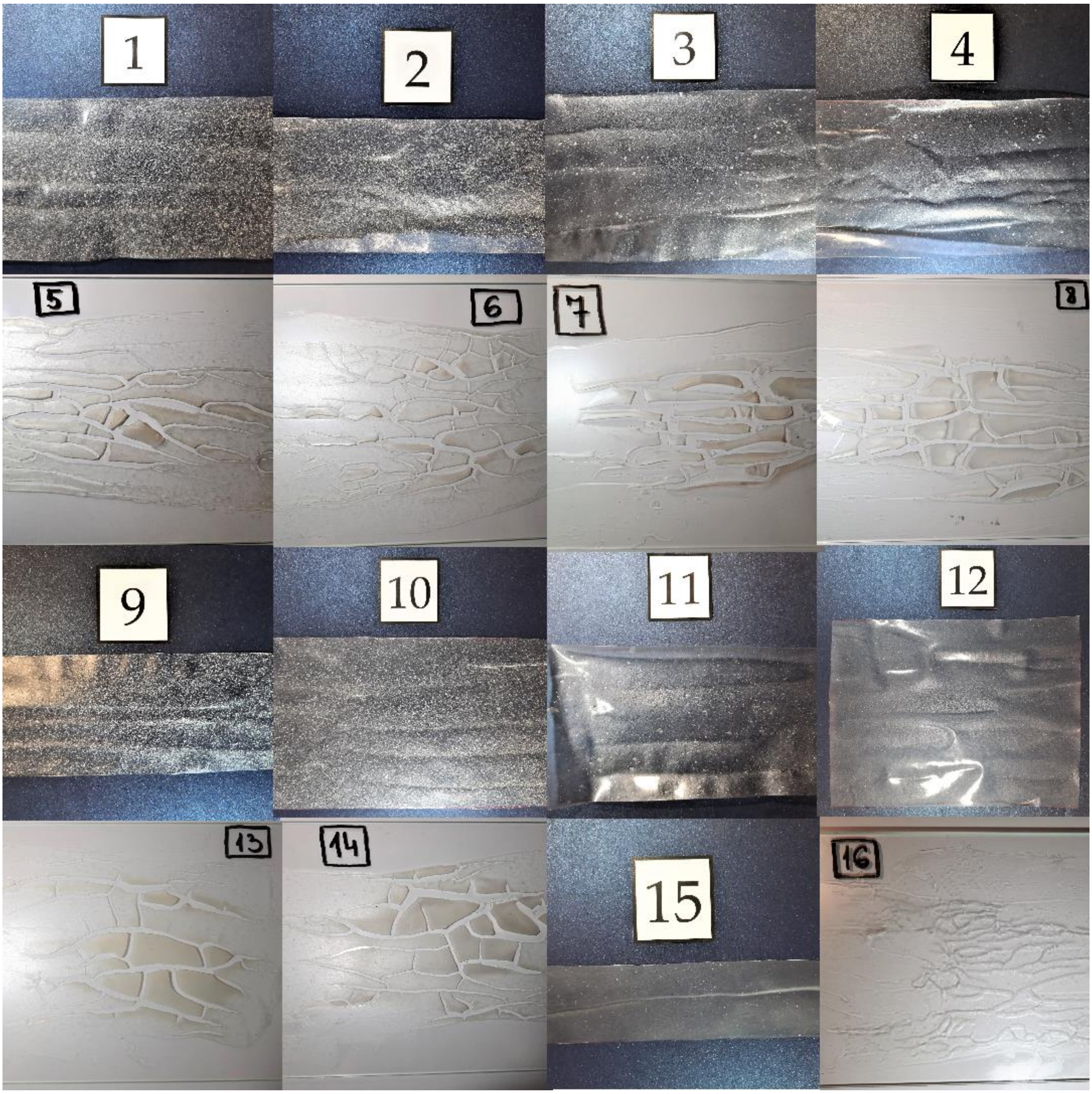

Fig. 15 Plant-based biodegradable plastic composites

2.4.2. Maximum force results

A maximum force test was performed on nine compositions (*No.1*, *No. 2*, *No. 3*, *No. 4*, *No. 9*, *No. 10*, *No. 11*, *No. 12*, *No. 15*. Load (*N*) and stress (*%*) were first measured, then the maximum force was calculated. The load was gradually increased. Due to the manual casting method, quite high errors were obtained. The results are inaccurate due to the different thicknesses of the samples. Therefore the manual film casting method is not suitable for the production of comparable films. Diagrams of load (*N*) and stress (*%*) results are presented in Fig. 16 – Fig. 20. The results directly depended on the different compositions of samples.

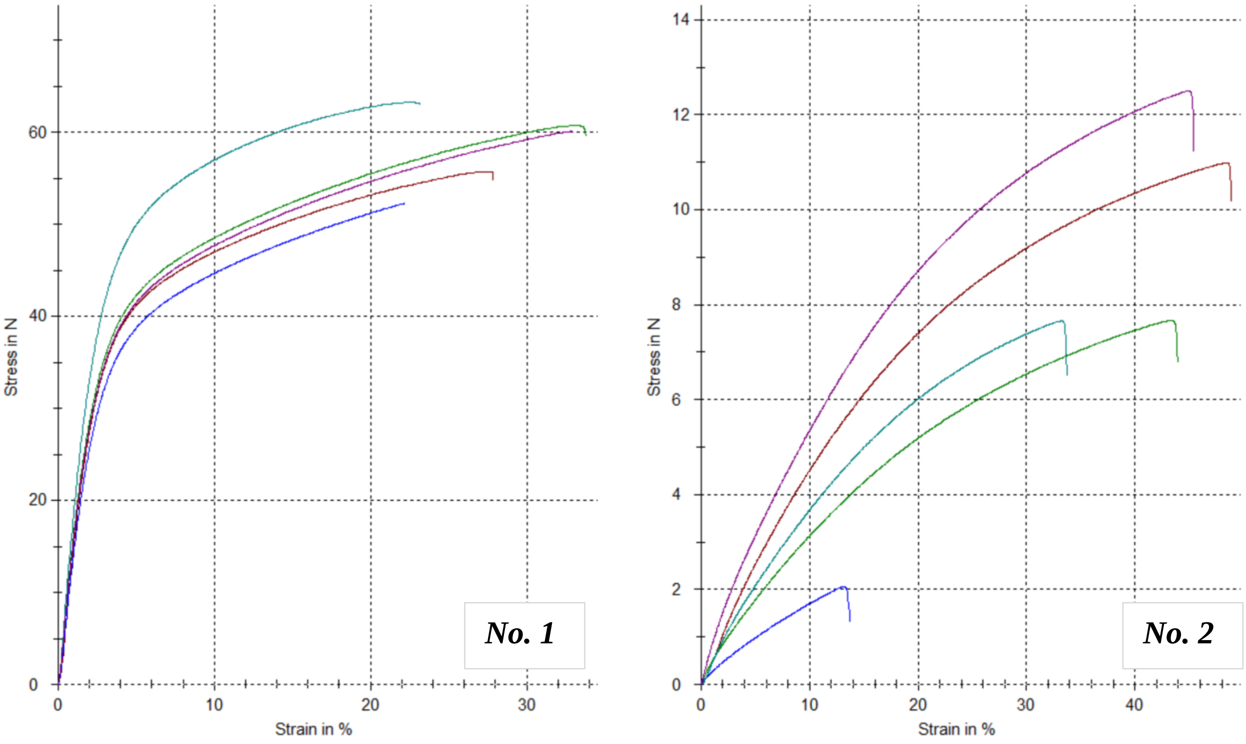

Fig. 16 Tensile stress and strain results of samples *No. 1* and *No. 2*

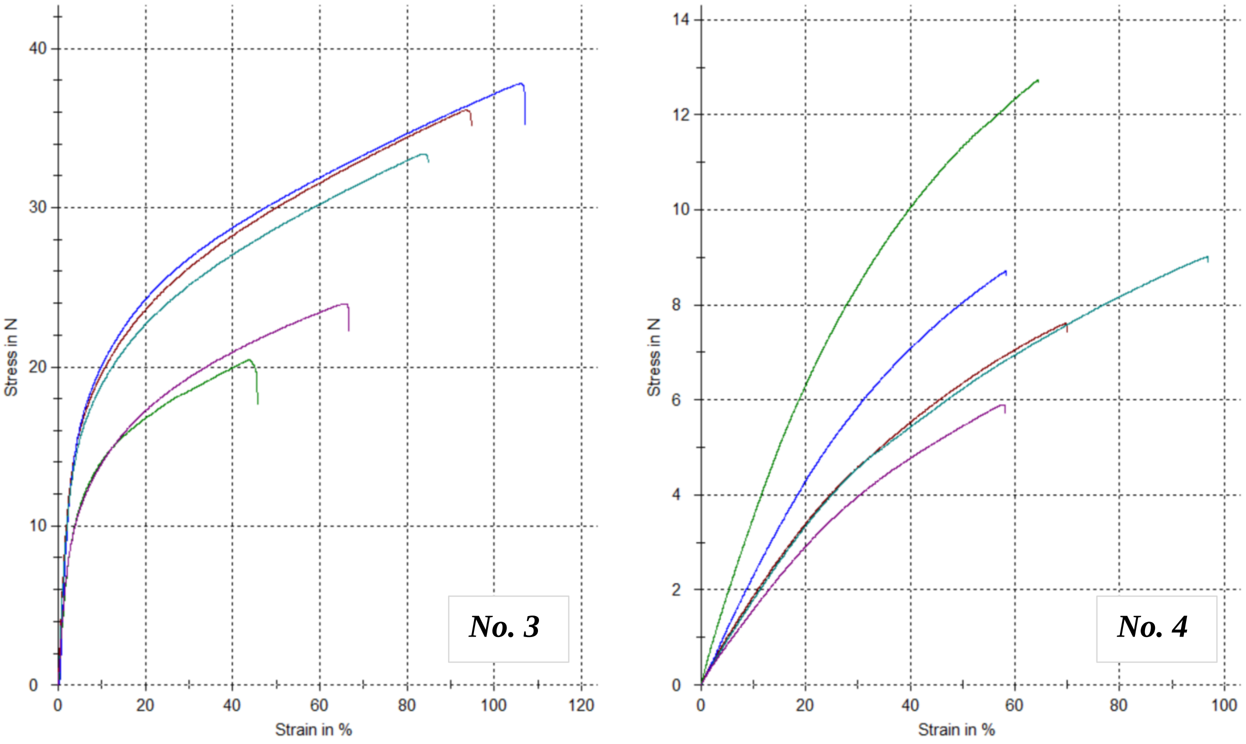

Fig. 17 Tensile stress and strain results of samples *No. 3* and *No. 4*

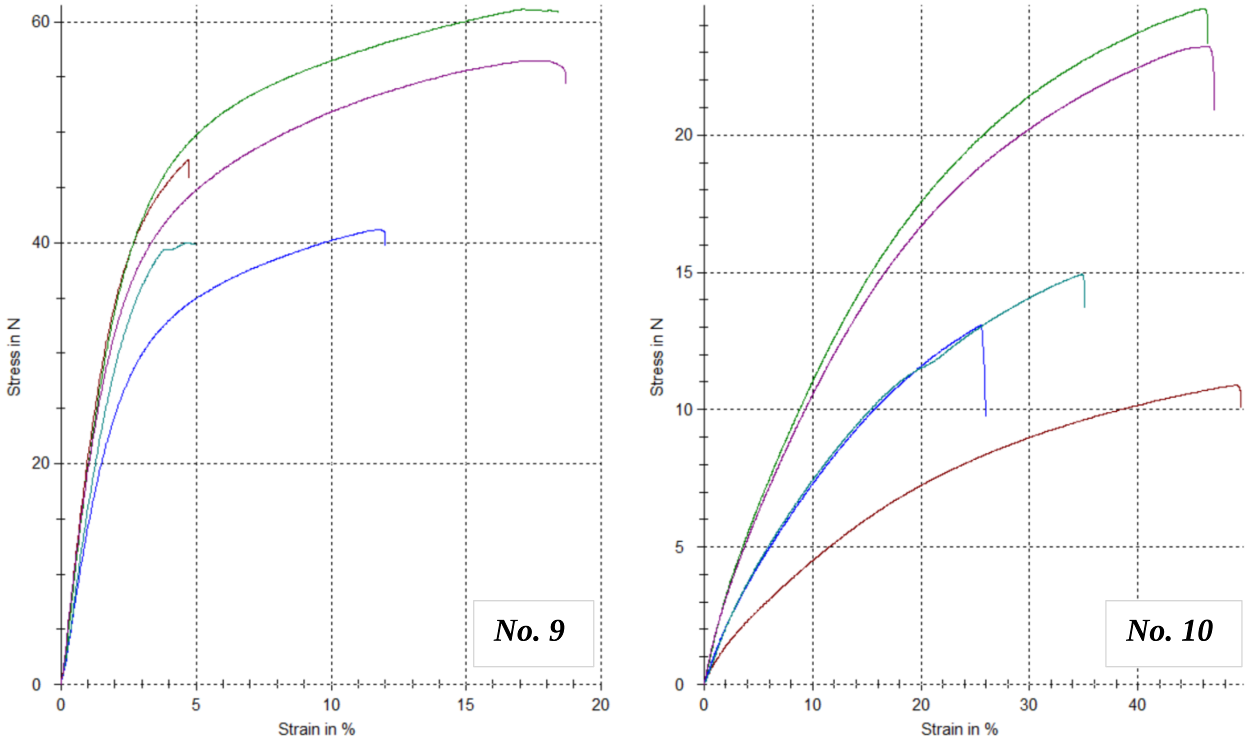

Fig. 18 Tensile stress and strain results of samples *No. 9* and *No. 10*

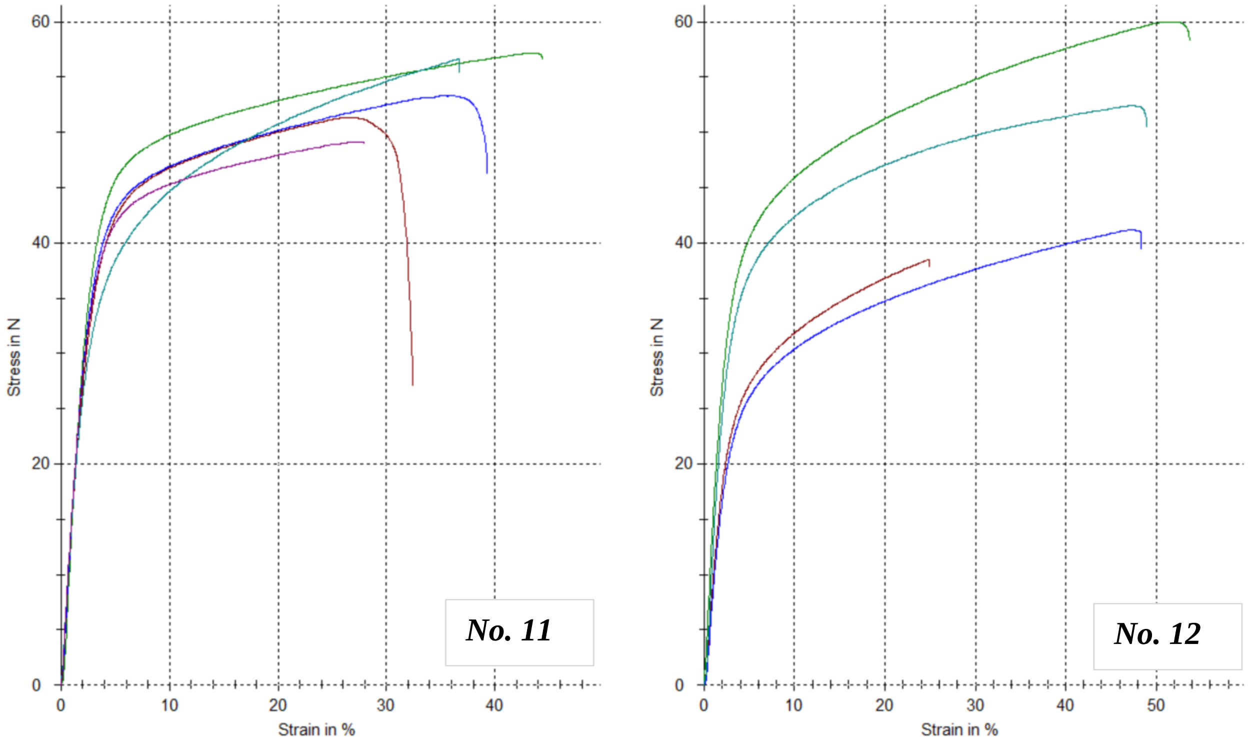

Fig. 19 Tensile stress and strain results of samples *No. 11* and *No. 12*

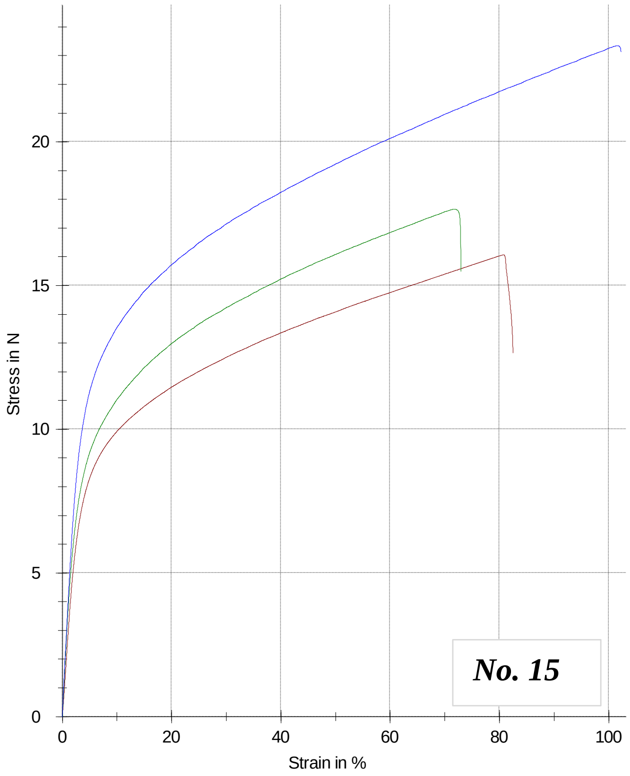

Fig. 20 Tensile stress and strain results of sample *No. 15*

Calculations were performed and summarized in Table 13. The values of maximum force (*N*), breaking force (*N*), percentage maximum force value (*%*), percentage breaking force value (*%*), and Young's modulus (*GPa*) are provided.

Table. 13 Mechanical properties of films

Composition	**FH, *N***	**FB, *N***	**εH, *%***	**εB, *%***	**E_{mod}, *GPa***
No. 1	58,45	57,91	27,66	27,99	1,009
No. 2	8,18	7,19	36,65	37,12	1,138
No. 3	30,33	28,57	78,74	79,83	1,062
No. 4	8,8	8,65	69,4	69,55	1,017
No. 9	49,28	48,11	11,21	11,78	1,024
No. 10	17,35	15,52	40,39	40,81	1,118
No. 11	53,54	46,83	33,84	36,15	1,143
No. 12	48,04	46,5	42,78	43,99	1,033
No. 15	19,02	17,08	84,71	85,89	1,114

FH**,** *N* – maximum force;
FB**,** *N* – breaking force;
εH**,** *%* – maximum force in percents;
εB**,** *%* – breaking force in percents;
E_{mod}, *GPa* – Young‘s modulus.

The highest value of the maximum force was determined in sample *No.1*, which consisted of potato starch, pea fibres, and higher sorbitol content. When evaluating the samples in the general context, the samples with the plasticizer sorbitol had the highest values. No direct effect of pea fibres was found. Maximum force data are provided in Fig. 21

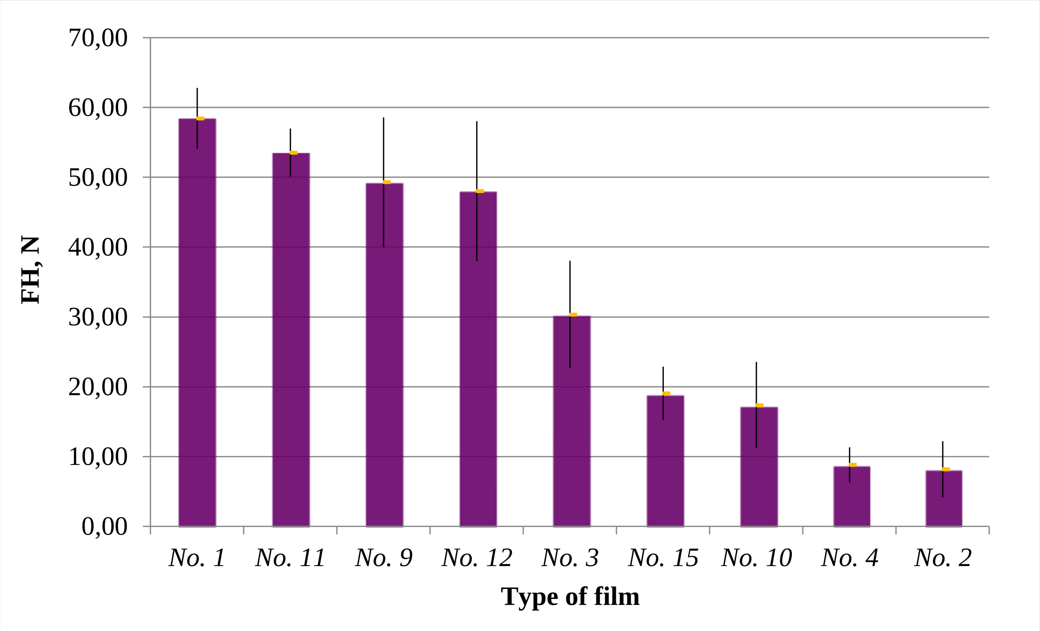

Fig. 21 Maximum force results comparison of the films

2.4.3. Young‘s modulus

When evaluating the values of Young‘s modulus, no repetitive cyclicity due to plasticizer or pea fibres were observed. Results range from 1,009 GPa to 1,143 GPa. The data are shown in Fig. 22. Comparing Young's modulus values of other plastics, the values of the samples designed in this work were closest to polypropylene copolymer:

- Polypropylene 1.00 – 1.40 GPa;
- Polylactic acid 3.80 GPa;
- Polyethylene terephthalate 2.80 – 3.50 GPa;
- Low-density polyethylene 0.11 – 0.45 GPa;
- High-density polyethylene 0.8 GPa [42].

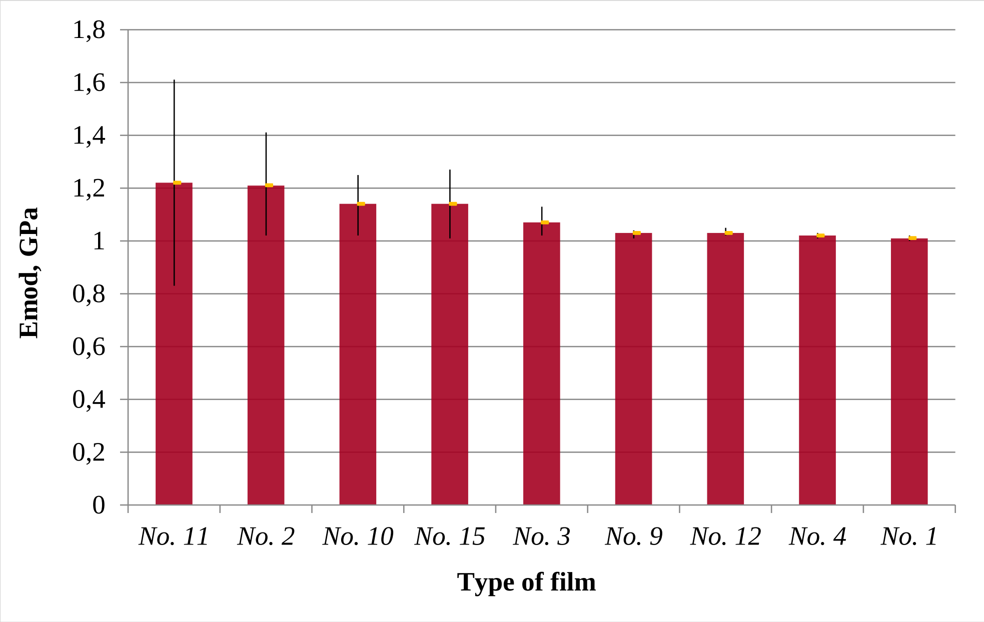

Fig. 22 Young‘s modulus of comparable films

2.4.4. Contact angle

Observing the data of the contact angle between the films and water, it was found that the films with pea fibres and plasticizer – glycerol had the highest contact angle values (*No. 2* = 38,7 °). Samples with the plasticizer sorbitol were the least resistant to moisture. An image of a drop of distilled water is provided in Fig 23. Numerical data are shown in Fig. 24.

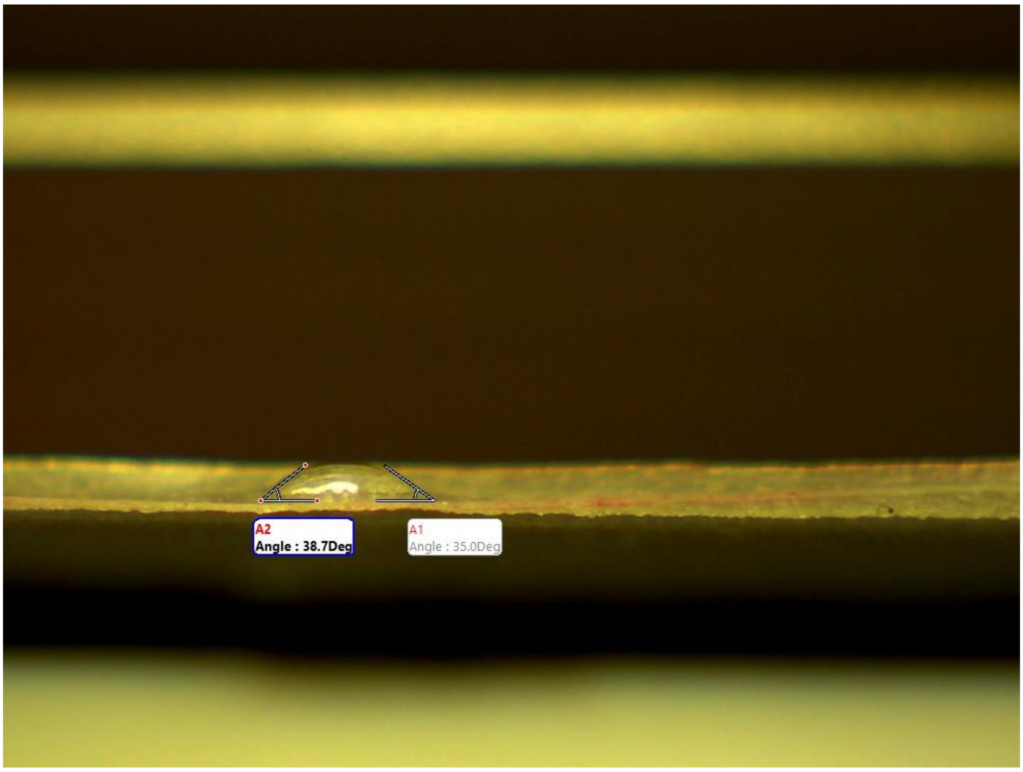

Fig. 23 Contact angle measurements of the film *No. 2*

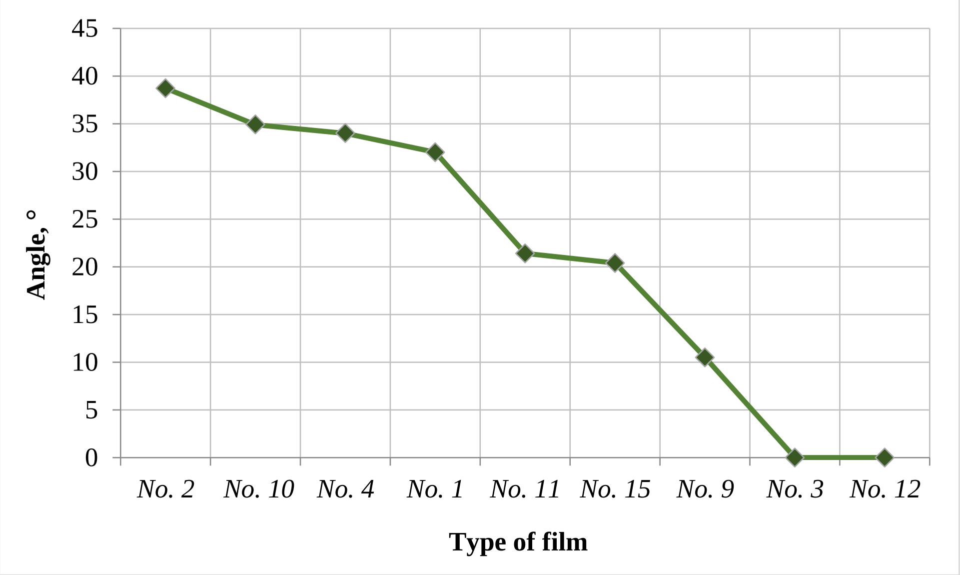

Fig. 24 Contact angle measurement results

The contact angle also indicates the level of surface treatment. Based on the requirements of ASTM D5946 (Table 9), it can be assumed that the machining level is very high (<71 °).

2.4.5. Thickness measurement

The thicknesses of the samples vary due to their composition, sheet compressive strength, and other influencing factors. This factor needs to be considered in concluding strength, as some samples are significantly thicker than others. The range varies from 0.24 mm to 1.41 mm. Thickness measurements are provided in Fig. 25. For accurate results, the thickness of all films should be the same.

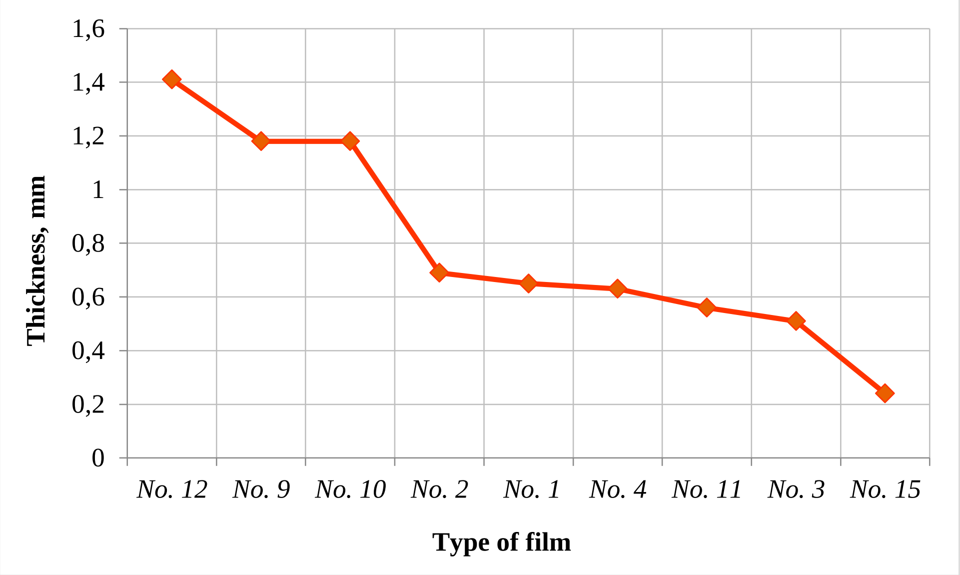

Fig. 25 Thickness measurements of films

For accurate results, the thickness of all films should be the same. For the production of a plant-based biodegradable bag, the film thickness should reach 0.05 - 0.0635 mm. Thus, it can be concluded that the samples produced are too thick for potential bag production. The excessive thickness of the film may have undesirable properties i.e. the plastic may become too non-rigid in structure, giving a false impression that it is strong. However, too hard plastic will not be flexible enough for a soft bag, and at low plasticity, the bag may break or crack due to too little bending. The thickness of the films identified did not depend on their composition but the method of production. Identical film thicknesses require the use of sandwich technology or single screw extrusion, where the film thickness can be tightly controlled.

2.4.6. Evaluation of oxygen formation by microscopic observation

Films were observed under a microscope *DinoCapture 2.0*. Although there were few oxygen gaps visible to the naked eye, the largest gaps were observed under the microscope that could lead to a further defect during the manufacturing process or when already using the packaging. The smooth surface of the film protects the product in the package, but there is a risk of losing barrier properties in the presence of oxygen gaps. The results are presented in Fig. 26 and Fig. 27. They can be affected by homogenization of the mixture during boiling, temperature regime, drying process, the strength of compression between sheets. In this case, the oxygen gaps formed due to the manual method of film casting, where oxygen remains between the sheets when the mass is compressed. The formation of oxygen gaps was influenced by the method of production, in this case, improper compression and drying. Generally, air gaps are not typical of the production of plant-derived biodegradable films and packaging therefrom. To solve this problem, special homogenizers that mix the mass evenly can be used, as well as dryers for this purpose. When evaluating the comparable films, the maximum air gaps were found in samples *No. 11* and *No. 12*. The smallest air gaps were determined in the comparative samples *No. 1* and *No. 3*, they were unified by the plasticizer sorbitol. Including single air gaps, the most even films were *No. 4* and *No. 15*. Therefore, it can be concluded that the films without coarsely ground pea fibres were the most uniform. When evaluating the further use of pea fibres in plant-based biodegradable films, the finest possible grinding fibres should be selected.

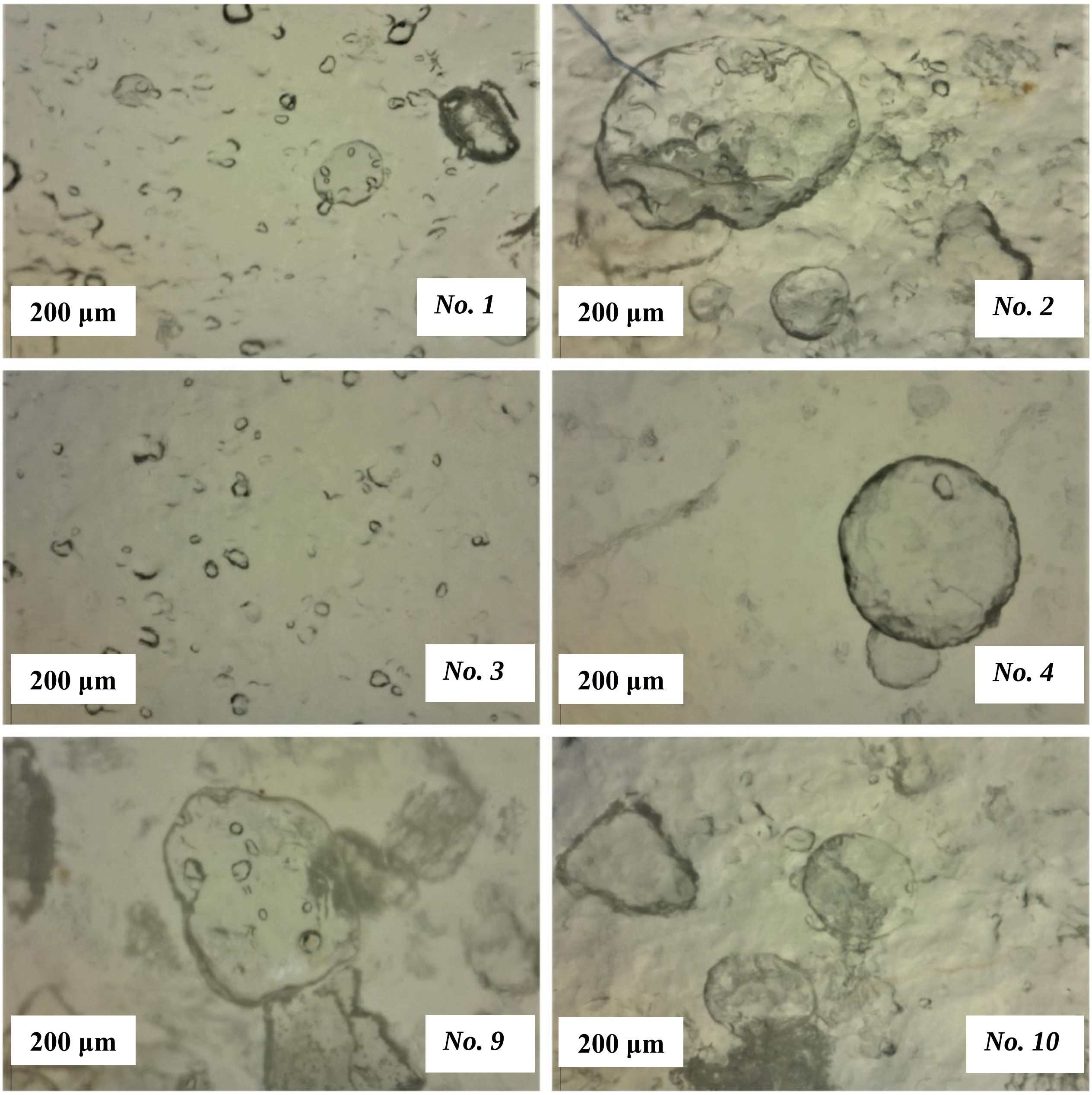

Fig. 26 Oxygen gaps in *No.1*, *No. 2*, *No. 3*, *No. 4*, *No. 9*, *No. 10* films

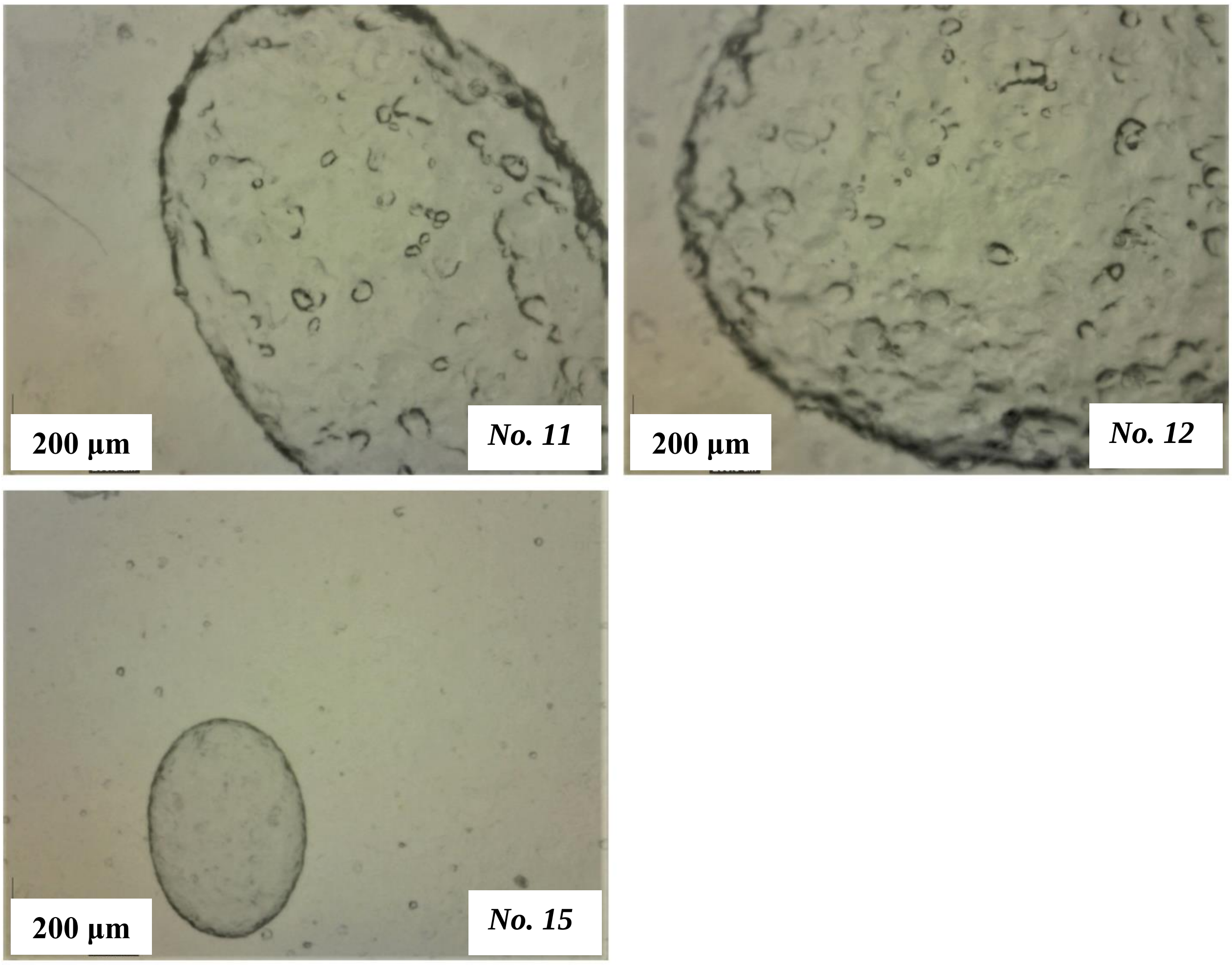

Fig. 27 Oxygen gaps in *No.11*, *No. 12*, *No. 15* films

Conclusions and recommendations

In the performed experiment, the visual and mechanical properties of comparable films were investigated. The result of maximum force under the action of different plasticizers and pea fibres fillers was evaluated. It was observed that the strongest films were composed of potato starch and sorbitol. The influence of pea fibres was distributed through both plasticizers. Among the different compositions, the distribution of Young‘s modulus was unequal, the regularity of the composition was not observed. Young‘s modulus values ranged from 1.009 GPa to 1.143 GPa. Therefore, compared to other types of plastics, the films designed in this work had similar values as polypropylene copolymers. After evaluating the contact angle data with water, it was found that the films made of pea fibres and glycerol were the most resistant to moisture, and the films made of the plasticizer sorbitol were the least resistant to moisture. Due to the manual method of film casting, quite large errors were obtained between the mechanical measurements, because the thicknesses of the films differed from each other, therefore the obtained results cannot be called very accurate. Also, due to the manual molding method, it was possible to detect oxygen gaps of various sizes in all films, which could affect the quality of the packaging. However, in real production, film casting is not used because a single-screw extruder is used, where the system is more closed and more accurate.

Continuing the idea of this study, nanocrystals made from natural potato or corn starch can be used to enhance the mechanical properties of the films. Nanocrystals are produced from starch granules by a hydrolysis reaction and the release of plates on a crystalline basis. This would increase the glass transition temperature of the elastomer matrix, the stress values before rupture. However, water absorption and decomposition temperature would also increase [43]. The most important parameter that needs to be improved in the films designed in this work is water resistance. If the film *No. 2* the highest value of the contact angle was 38.7°, the largest competitor in the production of plastic bags - polyethylene has the value of the contact of 96° [44]. Thus, starch-based plastic is more than twice as low as polyethylene in terms of water absorption. One of the many ways to improve water resistance is natural epoxy rubber. Epoxy natural rubber is obtained when rubber latex reacts with peroxy acid. This material has good elastic properties, so it is relevant for plant-based biodegradable films, not only for increasing water resistance but also for providing flexibility. Epoxy natural rubber provides resistance to fats and other sticky materials [45]. The films designed in this experiment were colourless, but for marketing purposes, the film should be painted. One of the choices could be natural pigments (curcumin, betalain, and others), but during oxidation, the dye begins to decompose and the film may fade. Therefore, colour fixers or synthetic paints are required. The possibility of printing could only be applied to films that do not contain pea fibres because due to the coarse grinding, the films have a coarse structure, which would make an even coating coverage difficult. Thus, the most appropriate way to give a visual appearance to a film is to add dye when the mass is homogenized (mixed).

3. Technological process of production

In the experimental part, reference films were produced in a laboratory way. When planning a real technological production path, it is important to choose the right equipment and its accessories to get the most efficient, cheap, and suitable product for the intended purpose. The following sections describe the production steps, the final product of which is a biodegradable bag of plant origin.

3.1. Production of biodegradable bags

When choosing a production method for biodegradable bags, it is important to take into account the main materials from which the product will be made. In this case, equipment capable of processing a high starch viscosity and ensuring a high processing pressure is required for the molding to be carried out continuously. There are two possible production methods:

a) First, produce biodegradable granules and then extrude them;
b) Using the main extruder, carry out the production without pellets and with dry starch.

Pellet production

In this work, it was chosen to produce pellets and make a plastic film from them. Granules are made from a mixture of potato starch, sorbitol, pea fibres, vinegar, distilled water. The pellets are produced in a size of 4 mm. Granular materials are substances with a particle diameter of more than 0.5 mm, and smaller particles are called powders. In most cases, granular products are superior to powder because they stay loose for longer and do not dust. During the granulation process, the monomers generate polymerization reactions. Granulation is the conversion of very fine powdered materials into granular ones, i. y. almost uniform agglomerated larger particles - granules, in interaction with the liquid phase. The best shape of the granules is spherical, because then they are mechanically stronger, less rubbed when poured, more evenly distributed during spreading. The granulation parameters of dry materials depend mainly on the moisture of the granulated mixture and the chemical nature or plasticity of the components [46]. The pellets are produced using a single screw extruder. Liquid and dry substances are poured through the funnel, they are formed, dried, and stored according to the chosen shape by heating.

Film production

The extrusion method is one of the main ones in the production of plastic bags because the polymers melt evenly at high temperatures at high rates. Such a method is suitable for extracting large quantities of identical packaging materials. This is especially true for polymers, as their low thermal conductivity and relatively high viscosity prevent easy and rapid dissolution in some volumetric containers during heating. The mass on the surfaces dissolves, but the mass on the inside remains unheated. In this case, temperature differences occur in the container and the heated and unheated masses are no longer mixed evenly. Thus, the extruder pumps the molten polymer, blowing to disperse the viscous mass, forming from it a thin film which is wound on rollers and thus twisted/renewed. A single screw extruder machine was chosen for this design line. The extruder consists of a hollow cylinder with heaters mounted on the outside. The screw is the main part of the extruder because according to its position and installation, the film will be formed. The propeller is driven by an electric motor through gearbox gears set to the required speed and power. Most of the energy is consumed by the engine to supply the molten mass. An example of a single screw extruder is shown in Fig. 28 [47].

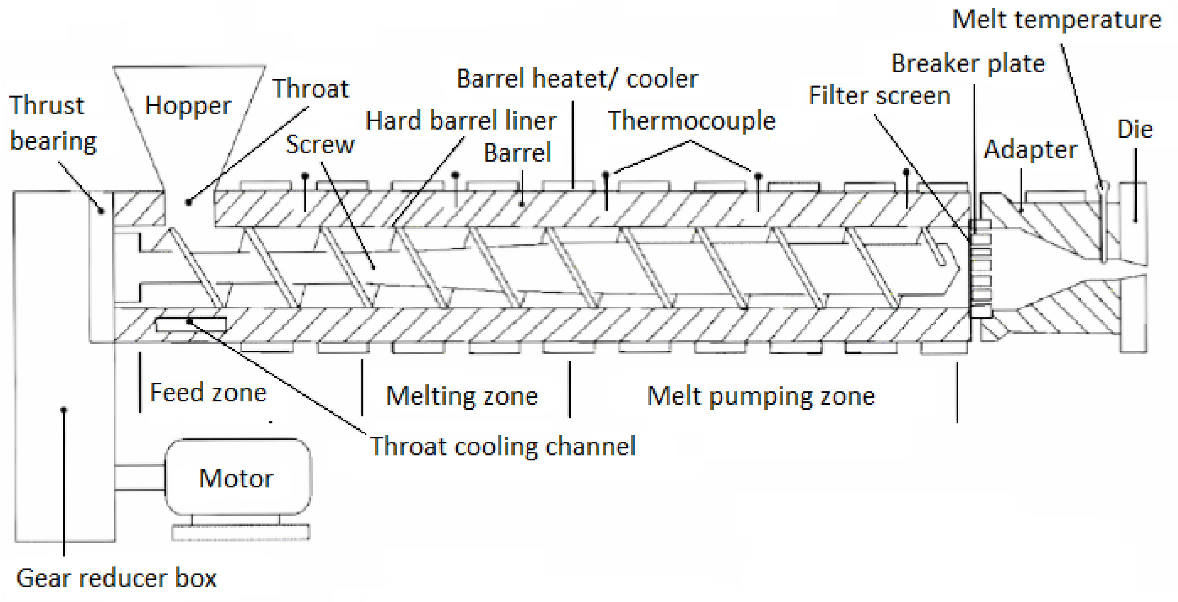

Fig. 28 Scheme of single screw extruder [47]

The extrusion method is one of the main ones in the production of plastic bags because the polymers melt evenly at high temperatures at high rates. Such a method is suitable for extracting large quantities of identical packaging materials. This is especially true for polymers, as their low thermal conductivity and relatively high viscosity prevent easy and rapid dissolution in some volumetric containers during heating. The mass on the surfaces dissolves, but the mass on the inside remains unheated. In this case, temperature differences occur in the container and the heated and unheated masses are no longer mixed evenly. Thus, the extruder pumps the molten polymer, blowing to disperse the viscous mass, forming from it a thin film which is wound on rollers and thus twisted/renewed. A single screw extruder machine was chosen for this design line. The extruder consists of a hollow cylinder with heaters mounted on the outside. The screw is the main part of the extruder because according to its position and installation, the film will be formed. The propeller is driven by an electric motor through gearbox gears set to the required speed and power. Most of the energy is consumed by the engine to supply the molten mass. An example of a single screw extruder is shown in Fig. 29. During mixing, pressure is created which pushes the mass to the place where the formation begins. Before forming, it is important to note that the screen filter built into the end of the extruder filters out foreign matter to prevent it from entering the finished film. The condition of the dissolved polymer determines the further properties of the finished film. Under the action of mechanical forces, the alloy is formed in a thin polymer layer sandwiched between the compressed solid pin and the surface of the heated container. The viscosity of the melt usually ranges from 50 to 1000 Pa · s. Large amounts of mechanical energy dissipate in the molten film between the firmly pressed stopper and the surface of the container. This process occurs when the motor rotates the propeller, pushing the polymer along the surface of the capacitive propeller. Mechanical energy during viscous decomposition becomes heat in a thin dissolved film and heats it, raising the temperature upwards. Increased thermal energy in the molten film is created by thermal conductivity, convection in a solid polymer stopper, capacitance, and melting mass. Excessively high temperatures or improper mechanical feeding can lead to oxidative, thermal, or mechanical degradation of the polymer [47].

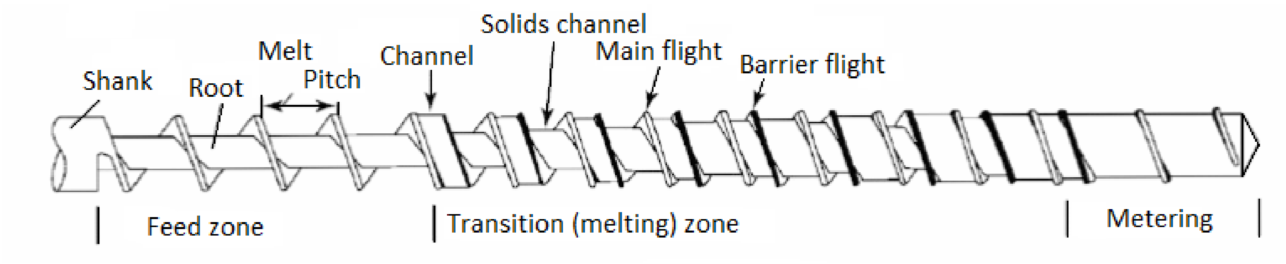

Fig. 29 Schematic view of the screw [47]

Blown film extrusion

Once the molten polymer has been filtered and travelled to the end of the extruder, an extrusion operation is initiated. In this process, the molten polymer exits the extruder head into a die. The liquid polymer flows around the axis and exits through a round annular hole to form a tube. Cool air is then blown through the die head into the ring. The cooling temperature is selected according to the airflow rate, the film pushing speed, and the temperature difference. The blown polymer forms into a tubular hollow bulge running vertically. Maintain a constant temperature (140 to 170 ° C). This tube is then levelled using rotating rollers and transferred to a wrapper so that the film is wound on a roll. In the blowing of the film, one of the main roles is played by the strength of the melt and its temperature, on which the strength of the bubble formed forms. The film blowing process involves the extrusion of a blowpipe, which is blown at an internal air pressure to thin the film as much as possible [48]. The thickness of most commonly produced films ranges from 5 mm to 200 mm [28]. After the cooling process, the inflated bubble is compressed by two small rollers that compress the film into two flat layers. With the help of rollers, the film is pulled away from the towers and further transported to cut (to get a rectangular shape and seal the cut space for another bag), making holes for the handles. The finished bags are folded and transferred to storage and sale. The film blowing process is shown in Fig. 30. Blowing the film has many advantages in making bags. Using the same stamp, it is possible to produce a film of different dimensions. No additional sealing of the sides of the bags is required. However, production requires very high premises due to the blowing upwards [31].

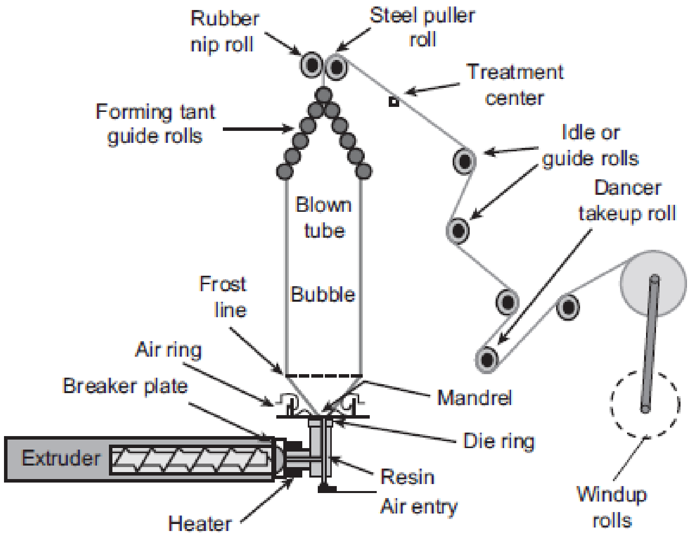

Fig. 30 Schematic view of the film blowing process [31]

Biodegradable bag production scheme includes:

- Storage and monitoring of raw materials;
- Mixing of raw materials;
- Pellet production;
- Storage of pellets;
- Automatic transport of pellets to a single screw extruder;
- Extrusion process;
- Film-forming (50 μm)
- Blown film extrusion;
- Rolling on rollers;
- Cutting into a rectangular shape (340 × 420 mm);
- Puncture of handle holes;
- Warehousing and disposal.

3.2. Price of raw materials

This section compares the prices of biodegradable raw materials. Table 14 shows the proportions and prices of primary raw materials. The materials used for the design of biodegradable bags are potato starch, sorbitol, pea fibres, distilled water, vinegar. Prices were calculated based on the materials used in the experiment.

Table. 14 Raw materials prices

Material	**Eur/kg**	**Eur/1000 kg**
Potato starch	1,1	1100
Sorbitol	1,1	1100
Pea fibres	1,60	1600
Distiliuotas vanduo	0,28	280
Actas	0,49	490
Total	**4,57**	**4570**

Thus, the price per tonne of the designed biodegradable mixture is 4570 euros. The pre-production raw material prices shown in the data provided mean that this is the cost of raw materials, excluding labour wages, electricity taxes, equipment depreciation, and other factors affecting the final price. Table 15 shows the prices of pellets already on the market. However, producers already on the market buy raw materials in large quantities, which allows them to obtain a lower price.

Table. 15 Comparison of pellet prices per tonne

Composition	**Tone price**
Potato starch, pea fibres (designed in this work)	4570 Eur
Modified Starch, PBAT, PLA	2742,45 - 3368,37 Eur [49]
Corn Starch, PLA, PBAT	3210 Eur [49]
Corn starch, PLA	4364,34 - 5211,53 Eur [49]

The prices of a ton of pellets are quite similar between those designed in this work and those already existing. This allows orientation as to whether the selected composition will be suitable for further bag production.

4. Composting, biodegradation, and recycling

Although the term "biodegradable" provides information on the original origin of the polymer, the term "biodegradable" describes the end of life of the polymer. Starch-based polymers do not accumulate because they are biodegradable to CO_2. The remaining materials are absorbed and returned by photosynthesis in the form of biomass. A polymer is called biodegradable when it can be metabolized by bacteria, yeast, or fungi. Metabolic processes can take place in the soil, compost, water. Biodegradable material is used by microorganisms as a source of energy and is converted over a period of time into biomass, water, CO_2, and sometimes, depending on environmental conditions, methane. In an oxygen-rich environment, aerobic decomposition takes place, the decomposition products of which become water and CO_2 [28].

4.1. Composting

Biodegradable plastics can be degraded anaerobically or aerobically. Used packaging can be decomposed in a home compost or industrial compost (in a specific container). Preference should be given to industrial compost (biogas production, anaerobic yeast composting), as special conditions (humidity, temperature) can be created in industrial compost, while at home the room temperature is usually 22-25 °C and the relative humidity is 40-50 %. Depending on the composition of the biodegradable plastic, higher temperatures and higher humidity may be required for faster degradation. To maximize the degradable effect, the co-disposal of biodegradable waste with municipal waste should be avoided, although this is not prohibited [50].

Anaerobic treatment / composting

It is a treatment method where complex biological, biochemical, and physical processes (microorganisms, soil animals, and their released enzymes) mineralize biodegradable waste, release nutrients and form humus. It is an organic part of the soil resulting from the decomposition of plant and animal residues. Usually, the soil contains 3-5 % humus. The main product of the anaerobic treatment process is compost, a completely decomposed (decomposed) mixture of organic waste used to improve soil properties. Compost undergoes the same processes as soil, but many times faster and more efficiently. Therefore, mature compost is not only a valuable fertilizer but also a promoter of soil vitality. For the anaerobic process to be successful in composting, the right conditions are necessary:

- *The moisture content* of compostable waste (from 70 % moisture, the anaerobic process begins, water fills the cavities). If biodegradable waste is not sufficiently moist, it should be irrigated. If not too wet, mix with dry biodegradable waste;
- *Carbon to nitrogen* (C: N) ratio (optimal decomposition begins when the carbon to nitrogen ratio ranges from 15: 1 to 30: 1);
- *Oxygen content in the process* (optimal amount 15-20 %);
- Process temperature (temperature values are shown in Table 16);
- *pH* (from 6 to 8);
- Biodegradable particle size (the finer the biodegradable material, the more it covers the surface area. This makes the materials more accessible to the decomposers but reduces the cavities for oxygen delivery to the mass [51].

Table. 16 Composting stages [51]

Stage	Temperature	Explication
Mesophilic stage	15 – 45 °C	Lasts 10 days (organic matter is broken down by mesophiles)
Thermophilic stage	50 – 75 °C	Lasts 3 - 4 weeks
Maturation (mesophilic) stage	45 – 20 °C	Lasts 1-2 months (mineralization of organic matter)

The main ways to treat compost are:

- *Cold compost preparation* – the compostable mass is used up to 2 times a year, the temperature inside rises to 40 ° C, the process takes up to 2 years);
- *Hot composting* – indoor home or open/closed industrial. The temperature rises to 70 °C, organic matter decomposes faster. Open industrial compost decomposes in 3 months, closed industrial compost in 20 days (not counting the maturation period, which lasts about 1-2 months);
- *Vermicompost* – compost is processed with the help of compost earthworms (Latin: *Eisenia fetida*), bio humus is obtained, duration 2 months [51].

To speed up the composting process, an open composting method is performed using a roof. Applicable mass reversers, air supply devices, living microorganisms that promote fermentation. Closed composting uses pipes or containers in which composting is intensified. Membrane films are used. For the films produced in this work, the most suitable method for composting is the aerobic hot method, because the films need to decompose at a higher temperature than those naturally present in the field [51].

EN 13432 Industrial composting

Biodegradable plastics must be internationally certified as compostable, according to certain indicators and tests performed. This standard specifies the minimum composition and degradation values that packaging must meet to be recycled by industrial composting. Plastic products certified according to *EN 13432* are marked with a special seedling shown in Fig. 31. This labelling helps consumers to become more visually quicker about the sustainability of packaging. Plastic products that are certified as compostable are sent to industrial composting recycling sites. Products that are not certified as compostable are mechanically processed [28]. Composting is the most appropriate method for treating biodegradable packaging after use. The compost remaining after the decomposition of the polymer can be used as a soil improver or fertilizer. High water retention capacity, rich soil conditions are real environmental benefits. This is especially true in soils suffering from drought problems. This method is suitable for very wet waste. The industrial composting method is divided into rot (active stage) and post-rot (hardening). The duration of the initial rot depends on the composting method chosen. Normally, microorganisms multiply on organic waste by breaking it down into carbon dioxide gas and water. Microorganisms use organic waste as nutrients and energy supplies. Some of the energy formed is released into the environment as heat generated. Organic waste is stored in piles during composting and, as a result, the temperature of the piles rises. As the temperature increases, microbial populations shift in the composting piles; microbes adapted to ambient temperature (mesophiles) stop working or even die and are replaced by microbes adapted to live at high temperatures (thermophiles) [28].

Fig. 31 Composting labels in different countries [28]

EN 13432: 2000 Compostable packaging
Composting processes are also described in standard EN 13432: 2000. This document describes that plastic can be called compostable when it decomposes to 90 % of its mass and mineralizes within three months, in industrial compost, which maintains a constant temperature (50-60 °C) [52].

4.2. Biodegradation

Biodegradation and compostability are not the same processes. Most polymeric materials are biodegradable in one way or another, but this can happen very slowly or quickly. Polymers based on starch have a very fast biodegradation time. The biodegradation of the polymer involves hydrolysis, oxidation, and photodegradation reactions. Also enzymatic decomposition of polymer chains into humus, water, carbon dioxide. The rate of biodegradation in the polymer is influenced by microbial characteristics such as type, distribution, pH, temperature, moisture content, oxygen, nutrients fed by the microbes. The rate of degradation is also influenced by the enzymes used by the microbes, which may be of the inner cell type or the outer cell type. The characteristics of the plastic that affect the rate of biodegradation are the hydrophilicity and hydrophobicity of the film surface, chemical structure, molecular weight, crystallinity. Biodegradation takes place in the following stages:

1. Abiotic reactions and intercellular effects of enzymes. Polymer chains are separated and shortened. Enzymes begin to break down the weakest parts of plastic. The crystallinity of the plastic is maximized;
2. The second stage occurs when the polymer chains reach a sufficiently small size, thus allowing them to be transported to the cells of the microorganisms, where they can be bioavailable and mineralized [52].

4.3. The problem of landfills

Landfilling is currently the most widely used method in the European Union. Biodegradable materials stay in landfills and decompose to form gas and filtrate. Landfill gas is made up of methane, which is one of the reasons for the growing greenhouse effect. Thus, it can be assumed that the landfill system is the most unsuitable for the breakdown of biodegradable packaging of plant origin. It is important to treat organic waste separately [28].

4.4. Recycling

Biodegradable packaging of plant origin that is suitable for recycling can be collected, sorted, and treated from unsuitable waste. Depending on the quality level of the plastic product, the type of recycling is selected. The schematic principle of recycling is shown in Fig. 32.

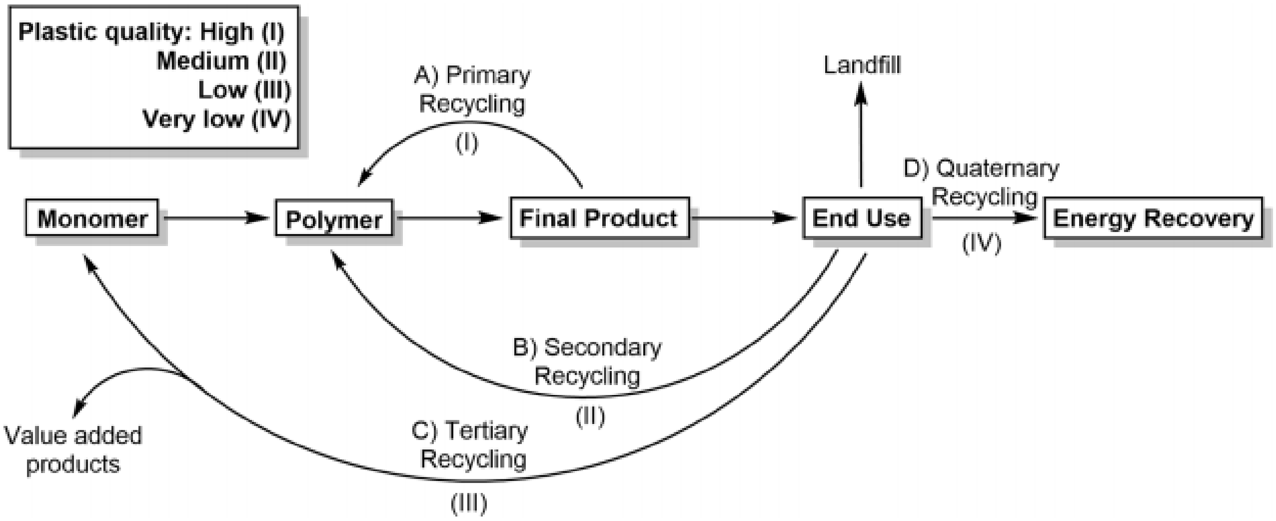

Fig. 32 Further use of plastic packaging in terms of plastic quality [52]

When assessing waste in terms of the quality of plastics, this should not be defined as a general quality indicator. Although this work designs environmentally friendly packaging, the following review of recycling methods in terms of plastic quality means that the plastic is mechanically durable [52]:

1. Closed-loop recycling, which can be carried out when plastic is classified as very high quality. When plastic is recycled in this way, it is again used to make another product or will be used as an ingredient in another composition;
2. Medium-quality plastics undergo mechanical recycling, which makes plastics less demanding and more suitable for further production. Blow molding, screw pressing, casting are used). This method does not contribute to increasing global warming (such as chemical recycling) and may therefore be the most appropriate of all recycling methods for environmentally friendly plant-based biodegradable packaging;
3. Chemical recycling is used when the plastic needs to be broken down into small monomers or other materials that can be used later. Depolymerization technology is used for these processes. With the help of chemical processing, it is possible to produce materials with enriched value. This means that polymers can be polymerized in unlimited quantities;

4. Energy recovery by incineration of low-quality plastic waste. Burning plastic generates large amounts of energy. The generated heat energy can be used for steam and electricity generation. This method of recycling should only be used in exceptional cases where other methods are not suitable to recycle the packaging. Noticeable negative effects are the loss of the molecular structure of polymers and the release of harmful chemical compounds that are incompatible with the idea of sustainable plastics [52]. Biodegradable plastics are usually co-incinerated with other conventional wastes, but incineration productivity is low due to the high moisture content (~60 %) of organic waste [28].

This section provided an overview of the main composting, biodegradation, and recycling methods. Starch-based plastics are most suitable for composting and biodegradation due to their sensitive structure, but can also be recycled depending on the possible composition.

4.5. Validation of results

The most important characteristics of biodegradable packaging of plant origin are discussed in the work. However, from a management perspective, it is important to analyze the strengths, opportunities, weaknesses, and threats. The results of the SWOT analysis are presented in Table 17.

Table. 17 SWOT analysis comparison

Strengths	Opportunities
• Easily degradable, compostable; • Recycling option; • No petroleum-based oil is used; • Starch is abundant in nature; • Promotes sustainability.	• Improvement of mechanical properties; • Greater water resistance is required; • A smaller fraction of pea fibres should be used; • Possibility to dye the film.
Weaknesses	**Threats**
• Low water resistance; • Higher price; • Films may not be suitable for printing.	• Customers may not be interested in the higher price; • Due to its sensitivity, the packaging may not consumer expectations.

Thus, taking into account the characteristics of SWOT, it can be concluded that plant-based biodegradable packaging will not lose its relevance, as the environmental plastic pollution increases uncontrollably every year. Therefore, sustainable solutions are becoming not only an alternative choice for conscious consumers but also a necessity to at least slightly slow down this complex problem worldwide. This type of packaging, depending on its composition, can be degraded, composted, or even recycled. Starch is an inexpensive raw material found in abundance in naturally grown vegetables. With the right marketing and information strategy, such packaging can lead to consumer satisfaction in terms of contributing to sustainability solutions. Biodegradable packaging should not have too low water resistance properties compared with daily used packaging, so it is necessary to improve it. Considering the weak points of this type of packaging, consumers could be dissatisfied with the higher price, but not with such good mechanical properties. Thus, in general, biodegradable plastic has the potential to be popular in the market, but in order for the price to be based on sustainability ideas, the mechanical properties must also be improved due to the performance of the main function. It is important to inform consumers that this is a short-lived packaging that decomposes in a short time and does not pollute the environment.

Recommendations

Plant-based biodegradable composite film compositions that were designed in this work were suitable for the study, but a more precise production method (sandwich method or single screw extrusion) should be chosen. Although pea fibres partially reinforced the structure of the composite film, smaller fraction fibres should be used to ensure that the samples are smooth, without a grainy surface in order to be able to perform printing processes. Sustainable packaging is labelled with dedicated standards, it is important that the idea of sustainability would be reflected on the packaging. Specific drying conditions should be chosen as some samples dried longer than others.

Conclusions

1. Plant-based biodegradable substances, their decomposition time, chemical and physical characteristics were developed. Potato starch and maize starch were used as the main raw materials for the production of comparative samples. Yellow pea fibres were selected to fill the polymeric structure. Sorbitol and glycerol liquids were selected as plasticizers. A total of 16 samples were prepared, 9 of which formed a suitable film for further investigation. 8 successful samples out of 9 were composed of potato starch;
2. Mechanical properties were evaluated. Samples with sorbitol plasticizer had the highest maximal force values. Young‘s modulus values showed no cyclicity between the compositions, but the values are close to polypropylene. The contact angle values were observed more than twice lower than the polyethylene contact angle values. Therefore, moisture resistance has been identified as the most demanding indicator of improvement. Natural epoxy rubber can be used to improve moisture resistance. Due to the manual molding method, the films were characterized by air gaps observed through a microscope, which may have a barrier-reducing effect on the package;
3. The technology of the production of plant-based biodegradable bags was discussed. The essential manufacturing processes required to make a bag were pellet production, single screw extrusion, and blown films extrusion. The cost of raw materials for the production of pellets was calculated. Pellets price compared to already existed on the market did not differ significantly;
4. The further utilization of the packaging has been assessed. The most suitable way to treat used packaging is composting and biodegradation, but depending on the composition of the polymer, recycling processes are also possible;
5. Plant-based biodegradable plastics composites designed in this work are a suitable and potential alternative for the production sustainable of packaging, but it is necessary to take into account the improvement of certain properties and to ensure the efficiency of the production process.

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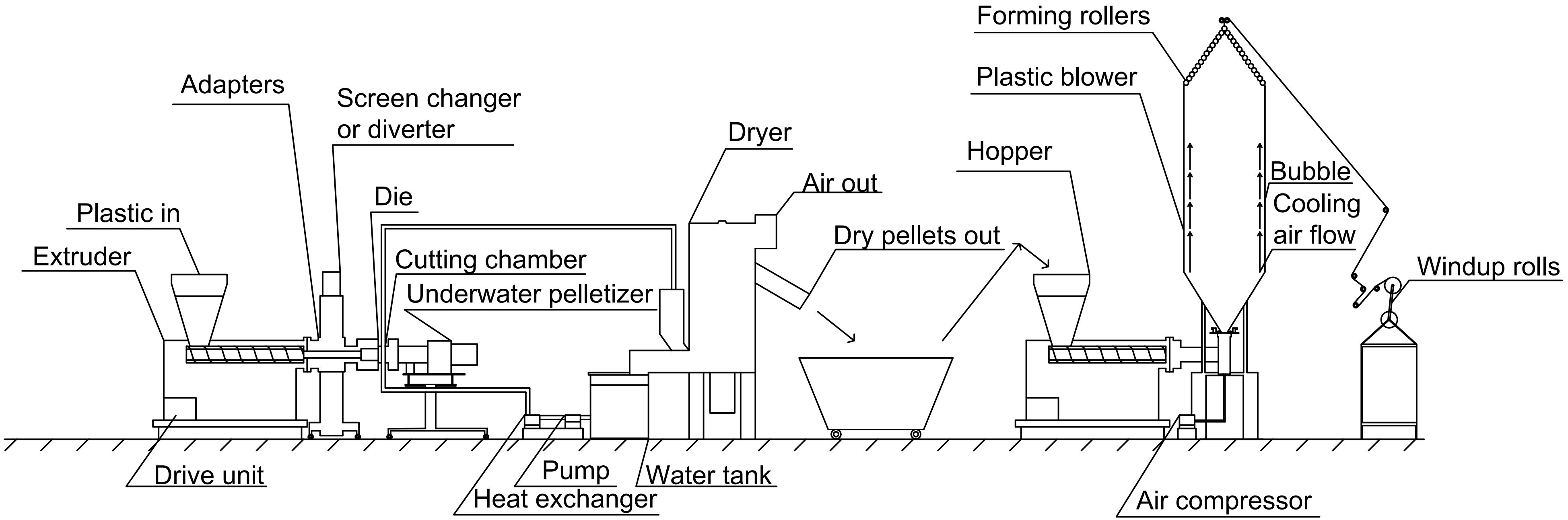
Appendixes

Appendix No. 1

Plant-based biodegradable plastic bags production scheme



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Plant-based biodegradable plastic composites for sustainable packaging

Lina Stabingytė[*]**, Laura Gegeckienė**[**]
** Kaunas University of Technology, Faculty of Mechanical Engineering and Design, Studentų str. 56, LT-51424 Kaunas, Lithuania, E-mail: lina.stabingyte@ktu.edu*
*** Kaunas University of Technology, Faculty of Mechanical Engineering and Design, Studentų str. 56, LT-51424 Kaunas, Lithuania, E-mail: laura.gegeckiene@ktu.lt*

1. Introduction

Recently, plastic products are being used in increasingly massive quantities. Cities around the world are expanding, population and urbanization is increasing, all of which is causing increased waste management challenges. If current waste generation levels remain the same, ~12 % of tons of plastic will be in the soil and environment by 2050. With plastics production growing at the same rate as today, the plastics industry could account for 20 % of global oil consumption [1]. The packaging industry ranks first in monitoring the production of plastic products by sector. Growth in plastic production may also be due to unavoidable reasons such as a global pandemic. The global COVID-19 pandemic that started in 2019 increased the amount of plastic waste even more (medical devices, additional food packaging) [2]. Due to the risks of petroleum-based plastics as an alternative, biodegradable plastics (BP) of plant origin are receiving increasing attention. *Novamont* (Italian company founded in 1989) biodegradable corn starch bag with patented Mater-Bi® technology is presented in Fig. 1. BP produced from biomass using the activity of microorganisms from themselves to degrade. This method of decomposition would significantly reduce the accumulation of plastic waste. Under the right conditions, BP decomposes into water, biomass and gas. The degradation level of BP depends on the chemical properties and functional groups (-COO-, -OH, -COOH) of the polymer, the chemical structure and the crystallinity of the polymer. Although this type of plastic decomposes easily in the soil and does not pollute it, the degradation process requires certain conditions that are not present in all environments [3]. The main aim of this work is to evaluate the mechanical properties of plant-based films of different compositions (with and without pea fibres) also the impact of plasticizers (sorbitol and glycerol).

Fig. 1 Biodegradable bag made from *Mater-Bi®*

2. Preparation of comparable plant-based biodegradable films

Films were made from potato starch, yellow pea kernel fibres, glycerol, sorbitol, distilled water. The mass was mixed with a digital mechanical stirrer *Onilab OS40-Pro* (set at 600 rpm) same time heated with an electric hotplate until 65° C was reached. Film casting process was done manually. The thick composite mass was applied to a sheet of glass using flattening spatula. Due to the sensitive structure, the sandwich method was not used in order to obtain the largest useful area. Drying performed for 48 hours, at 21-22 °C, relative humidity 45-55 %. The hardened films were carefully peeled off the sheets. Out of 16 samples produced, the results of 4 comparable films are selected to be presented. For each composition 5 samples were prepared. Compositions of films are presented in Table 1.

Table 1. Compositions of comparable plant-based biodegradable films

Composition	**Film sample**			
	No. 1	***No. 2***	***No. 3***	***No. 4***
H_2O	68,8	68,8	70,2	70,2
Vinegar (9%)	6,9	6,9	6,9	6,9
Potato starch	10,3	10,3	10,3	10,3
Pea fibre	1,4	1,4	-	-
Sorbitol	12,6	-	12,6	-
Glycerol	-	12,6	-	12,6
Total	*100 %*	*100 %*	*100 %*	*100 %*

3. Results

Due to the coarsely ground pea fibres, the films have a rough surface. Films without pea fibres were completely transparent. The view of starch based composite films is presented in Fig. 2.

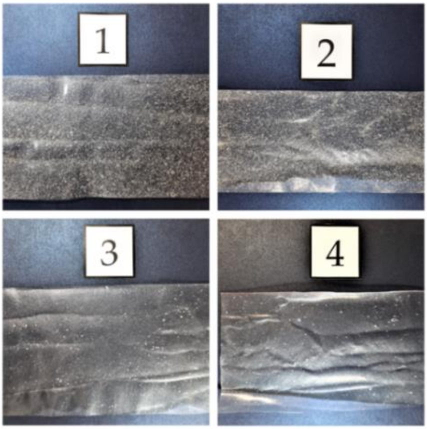

Fig. 2 Starch-based biodegradable plastic films of samples *No. 1* (pea fibre + sorbitol), *No. 2* (pea fibre + glycerol), *No. 3* (sorbitol), *No. 4* (glycerol)

The tensile force is the maximum stress that a material can withstand when stretched before the rupture. The maximum tensile force test shows that samples with sorbitol plasticizer (*No. 1* and *No. 3*) had higher values than those with glycerol. Sample *No. 1* = 58.45 N when sample *No. 3* (who has no pea fibres) = 30.33 N strength. Samples with glycerol (*No. 4* and *No. 2*) have maximum strength of 8.8 N and 8.18 N. Test were performed with *Zwick Z005* materials testing machine. The results are presented in Fig. 2.

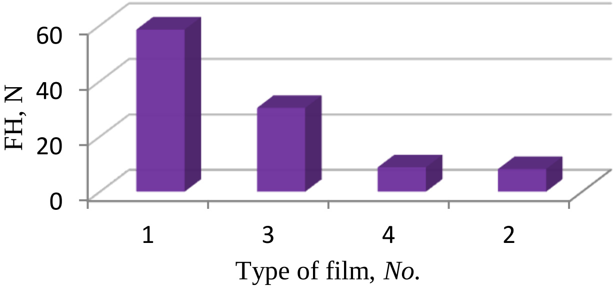

Fig. 3 Maximum tensile force comparison

Young’s modulus describes the compressive or tensile strength of a material. It is defined as the ratio of stress to relative elongation (deformation). The influence between compositions were not observed the results of Young’s modulus varies from 1.009 GPa to 1.138 GPa. In comparison, PP copolymer values are 1.00 – 1.40 GPa, PLA 3.80 GPa, PET 2.80 – 3.50 GPa, LDPE 0.11 - 0.45 GPa, HDPE 0.8 GPa [4]. Results are presented in Fig. 4.

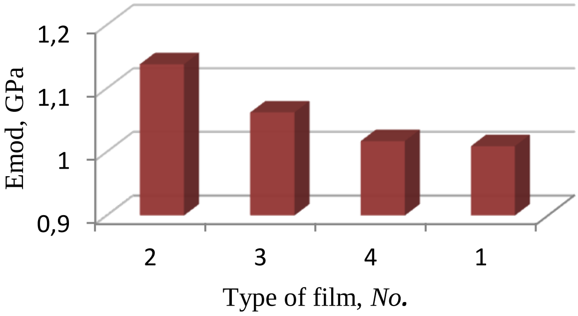

Fig. 4 Young’s modulus comparison

The contact angle is measured through a liquid, where the interaction of liquid and vapour meets a hard surface of film. The contact angle values shows the wettability of a solid film surface. Higher values of the contact angle were obtained in the samples with the plasticizer glycerol (*No. 2* = 38.7°, *No. 4* = 34°) . Sorbitol combined with pea fibres maintained a similarly high result (*No.1* = 32°), but the control sorbitol sample was not water-resistant (*No. 3* = 0°). The contact angle of a liquid on a surface test performed by S1008 method (*Karahan, 2007*). The results are presented in Fig. 5.

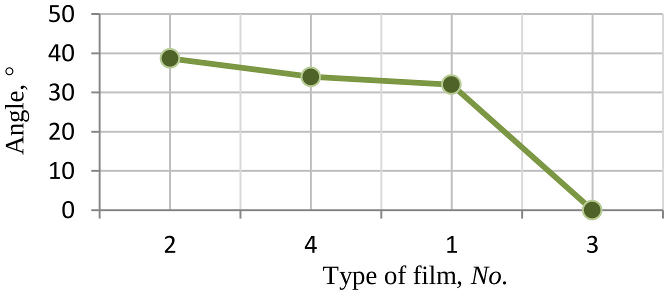

Fig. 5 Contact angle of a liquid on a surface

Films were observed under a microscope *DinoCapture 2.0*. Although there were few oxygen gaps visible to the naked, the largest gaps were observed under the microscope that could lead to a further defect during the manufacture process or when already using the packaging. The smooth surface of the film protects the product in the package, but there is a risk of losing barrier properties in the presence of oxygen gaps. They can be affected by homogenization of the mixture during boiling, temperature regime, drying process, strength of compression between sheets. In this case, the oxygen gaps formed due to the manual method of film casting, where oxygen remain between the sheets when the mass is compressed. The sample *No. 1* had small gaps of different diameters. The sample *No. 2* had unequal size, densely spaced oxygen gaps (the biggest – 0.8 mm length, 0.5 mm width). The sample *No. 3* depicts an even distribution of small oxygen gaps, when the sample *No. 4* has the smoothest surface, but one separate large (0.5 mm diameter) and one medium-sized (0.07 mm diameter) air gap. The results are presented in Fig. 6.

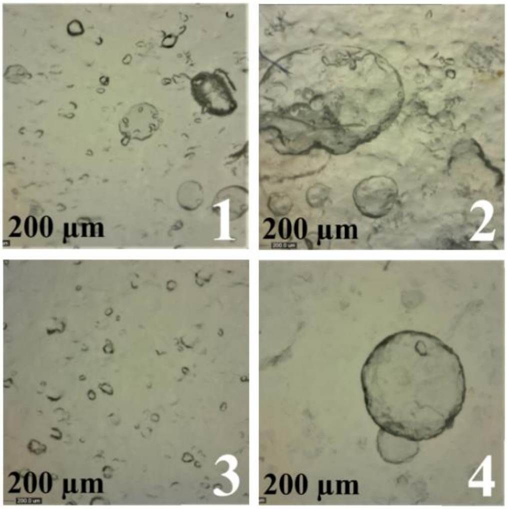

Fig. 6 Microscopic observation of potato starch comparable films *No. 1* (pea fibre + sorbitol), *No. 2* (pea fibre + glycerol), *No. 3* (sorbitol), *No. 4* (glycerol)

4. Conclusions

1. Films with yellow pea fibres visually had a rougher surface, films without pea fibres were completely transparent. Observation of the films through a microscope revealed that due to the manual casting method, the films have oxygen gaps, which can appear as barrier defects in the package.
2. The plasticizer sorbitol gives higher values for the maximum tensile

strength. Potato starch films combined with yellow pea fibres had a 48.11 % higher maximum stress than potato starch films with sorbitol alone.
3. Young’s modulus values varies from 1.009 GPa to 1.138 GPa.
4. Films with the plasticizer glycerol had the highest values of the contact angle. Therefore, they were the most resistant to moisture. Samples with yellow pea fibres and glycerol had 12.14 % higher values than samples with glycerol alone.

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Lina Stabingytė, Laura Gegeckienė

Summary

The aim of the article is to design comparable plant-based biodegradable composite films and to evaluate the mechanical properties for the purpose of application to the production of packaging (bags, wrapping films). An interaction effect of pea fibres and plasticizers (sorbitol and glycerol) on mechanical properties was compared. It was established that films with sorbitol as a plasticizer had higher maximum tensile stress values than films with glycerol. By reinforcing the film structure with pea fibres, they also increase the maximum tensile strength when combined with a sorbitol plasticizer. The highest values of moisture resistance were observed in the films with the plasticizer glycerol, and the combination with pea fibres further increases the resistance.

Keywords: biodegradable, film, plastic

Appendix No. 3

NETLOG 2021
International Conference on Network Enterprises & Logistics Management

CERTIFICADO

Lina Stabingytė

Has participated in the 2nd International Conference on Network Enterprises & Logistics Management – Logistics in the New Economy: Challenges and Opportunities in Supply Chain 4.0, which took place at Universidade Paulista, São Paulo, Brazil, from May 2nd to 5th, 2021.

Participou da 2nd International Conference on Network Enterprises & Logistics Management – Logistics in the New Economy: Challenges and Opportunities in Supply Chain 4.0, realizada na Universidade Paulista, São Paulo, Brazil, entre 2 à 5 de Maio de 2021.

São Paulo, maio de 2021

Alassane Ballé NDIAYE
Conference Chair

João Gilberto Mendes dos REIS
Conference Chair