Prospects for the use of plant materials in creating biodegradable eco-friendly packaging

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Abstract. This article provides information on the expansion of the base of vegetable raw material used in the packaging industry. During literature research the following criteria were considered: biodegradability, cheapness, availability, a sprouting area, an impact of used materials on condition of other industries. Plant-derived polysaccharides are contemplated as a source of biodegradable substances. The main subjects of research are plants with medicinal properties, invasive, technical and agricultural crops. The process for producing polysaccharide-based films was also examined. The possibility of introducing technologies into production was assessed. Conclusions are based on the impact of all stages of the process (collection, recycling, receiving and reusing) on the environment. Cogeneration technologies, pharmaceutical wastes and biotechnological biomass cultivation methods usage are taken into account to obtain efficient production.

1. Introduction
«Green chemistry» activity is in reducing or complete neutralization of contaminating materials or technologies in industrial and domestic practices. It also implies decreasing of environmental pollution connected with production or usage of certain materials. The importance of this scientific direction can be estimated considering all investments provided by governments of countries for research and development of alternative ways to generate and conserve energy, obtain biofuels and ecological materials. Some European countries are introducing standards, tax and subsidies system to stimulate development of environmentally safe technologies.

In this day and age, burning environmental issue is the use of polyethylene (in different forms as plastic wrap, polyethylene bag or packaging for food) and plastic. In natural environment polyethylene destruction time may vary from 100 years and over, required time depends on its type. It is also possible to reprocess plastic packaging with recleaning and smelting, but after processing this material is not sterile and cannot be used for storage and transportation of food or medical products. In fact, complete self-destruction of polyethylene and plastic is not possible.

One step in addressing the issue related to overproduction and overuse of non-biodegradable materials is using eco-friendly packaging produced from biodegradable polymer composites. This makes it possible to dispose material in a natural way without harming the environment. To create such material, it is viable to exploit plant-derived polysaccharides. The advantage of choosing polysaccharides lies in their non-toxicity, biodegradability and recoverability in nature with low carbon footprint. Further degradation products are also non-toxic to the environment.
The highest value in the manufacturing biodegradable packaging is related to the opportunity to use native plants of Ukraine, including weeds such as Arctium lappa L. and Cannabis ruderalis Janisch. It would help to optimize cost and quantity of material by eliminating transportation costs. Moreover, as a result of main active substances’ production from medical plant raw material pharmaceutical factories receive meal. There is a progressive idea of studying obtained meal in order to find out whether it contains any polysaccharides. In that way polluting emissions could be reduced and reusing of plant material would help to procure valuable packaging material. With the use of cogeneration technologies costs reducing and manufacturing of the closed type can be achieved [1].

2. Potential plants for use
The main material presented in this article is polysaccharides. Polysaccharides are complex carbohydrates that are polymers by their chemical structure. They are built from the remains of many thousands of monosaccharides and their derivatives combined by the polymerization reaction. By chemical structure features they are classified as homopolysaccharides and heteropolysaccharides. Homopolysaccharides are known as complex carbohydrates whose monomers are similar monosaccharides or their derivatives. Plant-based examples of such substances are starch, fiber, mucus, inulin and pectin. Heteropolysaccharides are complex carbohydrates that contain monomers with different chemical structures.

Synthesis of fructooligosaccharide-based composites film can be used in order to obtain completely biodegradable packaging. Inulin is a major component of those composites.

![Figure 1. Structural formula of inulin.](image)

List of plants and their parts with sufficient quantities of inulin:
- roots of Echinácea purpúrea contain up to 6%;
- roots of Cichórium contain up to 30% (both wild and cultivated types);
- Inula helenium L. contains up to 44% out of all chemical composition of plant;
- roots of Taraxacum officinale contain up to 24%;
- roots of Arctium lappa L. contain up to 45%;
- tubers of Heliánthus tuberósus contain up to 75%.

An additional source of suitable polysaccharides for sustainable packaging is plant raw material containing starch.
Figure 2. Structural formula of starch.

The most common sources are:
- Triticum vulgare grain (cultivated in 18 regions of Ukraine);
- Zea mays (cultivated for agricultural purposes);
- Solanum tuberosum (grown in large quantities throughout Ukraine);
- Platanthera bifolia (contains 27% of starch in tuber and listed in the Red data book of Ukraine, but cultivation is permitted on plantations for technical purposes).

Most of these plants are widely distributed throughout Ukraine. Therefore, there is no problem in getting raw material problems. Plants on the brink of extinction are allowed to be grown by modern biotechnology techniques. Our department scientists use in vitro biomass production methodology as an alternative source of secondary metabolites [2,3,4,5]. This method features some significant advantages over others: biomass grows under controlled conditions, the product is environmentally-friendly, technology does not dependent on climatic conditions, and, consequently, biomass is accumulated faster and year-round.

Application of cellulose fibrils instead of plastic is also a forward-looking development. Obtained carboxymethyl cellulose-based packaging can be fully composted.

Figure 3. Structural formula of cellulose.

List of promising supplies:
- Linum grain (a technical crop cultivated for industrial purposes);
- Cannabis ruderalis Janisch (permission for technical production is required at the moment, a wild variety can be used);
- roots of Cichórium (both wild and cultivated varieties).

It must be emphasized that the cost-effective solution would be to collect Taraxacum officinale, Arctium lappa L. and Cannabis ruderalis Janisch. In this way it is possible to produce eco-friendly goods from low-cost material and not to burden the bakery industry (exclude usage of wheat and corn). Since the above-mentioned varieties interfere with the planting of other plants, this can have a positive impact on non-invasive crops.
2.1. Market analysis of biodegradable packaging
The national economy of Ukraine is dominated by resource-intensive high-waste technologies and long-term lack of adequate response on burning ecological problems. As a result, the waste problem in Ukraine is of particular magnitude and importance. Significant use of resources, energy and commodity orientation of the national economy in combination with old-fashioned technological base defined extremely high rates of waste generation and accumulation. Mostly in that connection, The National Waste Management strategy in Ukraine until the year 2030 was adopted on 8th November 2017.

The strategy is aimed for ensuring the proper functioning of the national waste preventing, collecting, recycling and utilization system which also provide control of waste clearance and non-harming disposal [6].

Thus, for any enterprise to function properly, all resource saving activities must be applied and meet The National Waste Management strategy directions. One of those directions is biodegradable packaging creation and usage. At the moment, in Ukraine this niche is under development.

The table 1 provides a list of selected companies that exist on the packaging market.

| The company name | Trademark | Description |
|------------------|-----------|-------------|
| “Lamagrain” (France) | Biolice | PLA derived from corn Bags, net, trays, Q-tip tubes, compact disc boxes |
| “NatureWorks” LLC (the USA) | Ingeo | PLA derived from corn Bags, trays, cups, bottles (PET analogues), polystyrene analogue, disposable utensils |
| “TyssenKrupp” (Germany) | PLA obtained from corn Bags, net, linings, disposable utensils | “TyssenKrupp” (Germany) |
| “Nowamont” (Italy) | MATER-BI | Plant-based starch Bags, net, linings, disposable utensils |
| “SPHERE” (France) | Plant-based starch Bags, wraps | “SPHERE” (France) |
| “Plantic” (Австралія) | Eco-Plastic | Plant-based starch Bags, linings, disposable utensils |
| “BASF” (Germany) | Ecoflex, Ecovio | Plant-based starch Bags, net, cups |
| “Innovia” (the USA) | Nature Flex | Cellulose Film materials for packaging |
| Ukraine | ECOMAIZING | Biodegradable bags, corn starch and completely biodegradable plant-based materials |
| Ukraine | RE-leaf | Fallen leaves Wrapping paper |

Therefore, biodegradable packaging can be divided into three groups:
- synthetically produced materials;
- natural polymer-based materials obtained by biological modification;
- additives that enable synthetic polymers to degrade (dangerous components) [7].
3. Features of creating material

Cichórium has great potential for valorization in view of the fact that its roots are rich in inulin and cellulose. The plants can be used for obtaining cellulose fibrils.

Despite the existing technology of plastic film manufacturing from cellulose by fatty acylation, scientists examined obtaining film by chemical acylation of inulin with methacrylic and lauric chains [8]. A scheme and proportion of successful experiment were obtained after many failed experiments. These failures were caused by difference in reactions of inulin and cellulose with lauroyl chloride (composition plasticizer).

High-quality plastic films are formed in result of dissolved inulin in 4-Dimethylaminopyridine (DMAP) and subsequently added equimolar ratio of methacryloyl and lauroyl chlorides. The reaction is heated at 80°C for 2 hours. Sediment dissolution is performed using methanol and chloroform. The final stage is to form film - obtained product is dissolved in dichloromethane with followed evaporation of the solvent at room temperature.

The same technology makes it possible to obtain biodegradable plastic film not only from Cichórium, but from such plants as Echinácea purpúrea, Inula helenium L., Taraxacum officinale and Arctium lappa L.

Biodegradable thermoplastic can be also produced from starch. Nowadays, a range of starch-based polymer composites was commercialized in the USA and some European countries. Products from these materials are concentrated on eco-friendly markets. The ability to use starch greatly reduces the cost of making the material.

Basic technology of creating thermoplastic involves destroying starch granules. For this, processed in extruder starch is stabilized with cold water or plasticizers (phenol, glycerin) and heated. After being heated, starch granules break down and melt to highly viscous liquid. Hydrofoil polymers (polyvinyl alcohol, polycaprolactone, polyoxyethylene, polyvinylpyrrolidone) are added to the obtained liquid to stabilize an amorphous film structure by formation of hydrogen bonds with starch itself [9].

In Ukraine starch for creating thermoplastic can be obtained from Triticum vulgare, Zea mays and Solanum tuberosum. Triticum vulgare has the greatest potential for use due to its high yield in many regions of Ukraine.

In the article dated 25th September 2020, Croatian scientists V O Bulatovic, V Mandic, D K Grgic and A Ivancic investigated thermoplastic starch and polylactic acid composition for gaining complete biodegradability, fast degradation and obtaining economically viable material [10].

One of the most widely used components for eco-plastic is cellulose. Biodegradable plastic can be derived from hydroplastic cellulose polymers and cellulose acetate. Cellulose-based composites are the first-ever source for obtaining completely transparent film. The film was created by Lithuanian scientists in Kaunas University of Technology in 2019 to meet the needs of the food industry and replace polyethylene food packaging [11].

In Ukraine cellulose can be derived from wild variety of Cichórium, Linum grain and even weed plant as Cannabis ruderalis Janisch that grows throughout Ukraine. As an example, it is advisable to consider the most common methodic of creating sustainable packaging from cellulose acetate. Powdered cellulose acetate is mixed on high speed with low molecular weight liquid and oligomeric substances in proportion 2:3 or 1:3 (supplements are non-toxic). Obtained powder is granulated in extruding machine and dried for 2 hours. At the final stage material is processed by injection molding, then by extrusion blow molding and formed by pressing sheets to form final product.

Carboxymethyl cellulose-based biodegradable material is well-known for its costliness, stability and relatively short decomposition time.

In the research dated 15th December 2020, Indian, Malaysian and Vietnamese scientists suggested ways to reduce the cost of material by using their country’s distinctive agricultural wastes as sugarcane bagasse [12]. In the long run, it is possible to obtain same results from a sugar mill that cultivated in Ukraine. Waste from its technological usage is commonly granted on animal’s food additives or utilized.
There are few steps to produce packaging. First of all, material is prepared by drying agricultural waste in the oven at 80°C and processed for obtaining cellulose pulp. That pulp is dried again and filtered with laboratory mesh. For complete lignin removing it is handled with sodium chloride and acetic acid. Received mixture is heated under a water bath and later filtered. Obtained earlier cellulose is added to mixture of sodium hydroxide and isopropanol under a water bath to produce carboxymethyl cellulose (for cellulose alkalization). The material then is etherificated with addition of sodium monochloroacetate for 3 hours, filtered and resuspended in methanol. The mixture is repeatedly filtered and washed in methanol in order to neutralize sodium hydroxide residue. At the last stage methanol residues are removed.

Packaging is produced of carboxymethyl cellulose, gelatin and agar composite. Obtained material is molded in shape and dried for one day. The finished polymer film is removed with a sterile blade.

4. Conclusions
As a result of the literature research, main focus was on such indicators as:
- availability;
- cheapness;
- positive impact on the environment;
- closed-cycle manufacturing capacity;
- industrial wastes research.

An important fact is that it is possible to use invasive plant crops as a source of polysaccharides. This can be a smart practical environmental solution to deal with ecological issues.

The expansion of the biopolymer-based packaging market addresses such issues as:
- avoid environmental contamination owing to biopolymers degradability in natural conditions into safe components over a period of weeks to months;
- use of biopolymer packaging materials is safe for human health;
- source for the raw material to obtain eco-packaging are plant crops, so scarce oil reservoirs and unstable oil prices are important reasons for polymers production to switch to recoverable raw materials;
- this type of production does not require any special equipment and carried out according to traditional technology.

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