Developing the Composition of Biodegradable Film for Food Packaging

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Abstract. Searching for new food packaging material that would have neutral physical and chemical properties, would not be toxic, and would be degradable is a relevant problem for contemporary science. Biodegradable polymers are usually produced by polymerizing biological raw materials. This property of new materials will help solve the problem of waste. The authors produced an elastic film that does not crack when bent, recovers its original shape when unbent, can take virtually any shape, and has no smell and a neutral taste. The authors tried to assess the degradation of films in the natural environment. The film produced can be used for the manufacturing of biodegradable packaging for the food industry.

1. Introduction
Over the last decade, the problem of solid household waste disposal has got special importance. Only in Russia, up to 200 million cubic meters of solid household waste is generated every year, and more than half of it is food packaging. Biopolymers that can degrade under chemical, physical and biological impact are developed as an alternative to classic polymers. Biodegradation is the process of complex substance disintegration due to various impacts. These mostly include air oxygen, heat, and sunlight, as well as biological microorganisms, such as yeast, fungi, etc. These factors can work alone or combined, leading in the end to material fragmentation due to molecule disintegration into low-molecular compounds that take part in the natural cycle of matter.

2. Relevance and research objectives
Searching for new food packaging material that would have neutral physical and chemical properties, would not be toxic, and would be degradable is a relevant and pressing problem. [1]

3. Review
Currently, there are food packaging films in use, comprising such organic compounds as gelatin, glycerin, propylene glycol, etc. The literature describes using gelatin for the production of biodegradable films. It is a protein product, containing polypeptides with various molecular mass and able to generate aggregates of the high molecular mass of 300000. Gelatin has no taste or smell and it
can expand in water. Gelatin contains about 18 amino acids. The amino acid profile of gelatin is shown in Table 1. [2]

Table 1. Gelatin amino acid profile.

| Essential amino acids        | Per 100 g of gelatin | Nonessential amino acids | Per 100 g of gelatin |
|------------------------------|----------------------|--------------------------|----------------------|
| Leucine (BCAA)               | 2.64 g               | Arginine                 | 6.62 g               |
| Isoleucine (BCAA)            | 1.23 g               | Histidine                | 0.66 g               |
| Valine (BCAA)                | 1.93 g               | Glycine                  | 19.05 g              |
| Lysine                       | 3.46 g               | Aspartic acid            | 5.27 g               |
| Threonine                    | 1.48 g               | Glutaminic acid          | 8.75 g               |
| Tryptophan                   | 0 g                  | Alanine                  | 8.01 g               |
| Methionine + cysteine        | 0.61 g               | Proline                  | 12.3 g               |
| Phenylalanine + tyrosine     | 2.04 g               | Serine                   | 2.61 g               |

Gelatin can expand in water at room temperature, and if heated up to over 40°C, it forms gels that have a spiral configuration. The gel formation mechanism is connected to the formation of a threedimensional network structure. When the gel is cooled down, polypeptide chains comprised of the amino acid parts take spiral configuration that is stabilized by hydrogen bonds. These bonds are distributed along the entire chain, which explains the calorific reversibility of gelatin gels. Unlike some polysaccharides that are also capable of gel formation, gelatin gel formation does not depend on the pH and does not require other reactants, e.g. sugars, salts or divalent cations [3].

The authors [4] produced an edible film from gelatin, glycerin and olive oil, the structure and durability properties of which were studied. The durability and elasticity of the film increase along with the increase of olive oil proportion.

Andreuccetti C., Carvalho R.A., Grosso C.R.F. tried to inject hydrophobic plasticizers made of citric acid and soy lecithin in the edible gelatin-based film [5]. After durability and rupture-resistance tests, it was ascertained that the amount of hydrophobic plasticizers does not influence the elasticity of the end product.

Recent years show growing interest to propylene glycol as one of the cheapest plasticizers for the production of biodegradable compositions. Propylene glycol is extensively used in the food industry as a thickening agent and a stabilizer to achieve the necessary viscosity and soft structure of the product. The main criteria of using propylene glycol as a plasticizer are its capability to compound immiscible ingredients and prolong shelf life, its availability and low price. Plasticizer molecules reduce the attractive forces between protein molecules at low humidity, making the film more flexible and elastic. However, if an excessive amount of the plasticizer is used during film production, its moisture-binding capability and durability will deteriorate.

Eco-friendly natural compounds can be an alternative for the organic components of films. Enzymes are biological catalyzers that can affect substrates selectively and decompose them. Substrates can be represented by high-molecular natural compositions, e.g. cellulose, proteins, and other natural polymers. On the other hand, there is a growing interest in the transglutaminase enzyme, which does not destroy the substrate but binds protein molecules in a solid structure.

Natural transglutaminases in various forms can be found in microorganisms [6], in crustaceans and plants [7, 8, 9], in vertebrates [10, 11] including people [12]. Since transglutaminase is involved in the metabolic processes of virtually all living beings, there are methods for excreting this enzyme from mammals. However, the amount of the enzyme obtained this way is not worth the inputs, so currently, there are people working on excreting transglutaminases from plants [13].

Transglutaminase can form a protein structure that becomes stable in various temperature intervals and at different pH values. It is due to the fact that the enzyme binds with the substrate via covalent
links of the amino groups of different proteins, thus producing a durable structure that would be resilient to mechanical impact. Besides, its stability does not depend on the other components of the environment, and the protein matrix formed by the enzyme is similar to the natural structure of protein tissue [14, 15]. These factors provide for absolute eco-friendliness of end products.

4. Methods and materials
In this research, the BioBond TG–EB3 transglutaminase enzyme from Flora Ingredients was used. Temperature activity range of the transglutaminase is between 10 and 55°C, the desirable pH is from 6 to 7. Transglutaminase inactivation can be observed as a result of temperature action, the acidity of the environment and the duration of oxygen exposure. The enzyme disintegrates completely when exposed to the temperature of 72–75°C for 5–10 minutes [16].

The reaction of transglutaminase and the substrate protein in various proportions is determined by the presence and availability of the necessary amino acids in gelatin, as well as the factors promoting enzyme activity (pH value, fermentation time, temperature). As a result of this reaction, covalent cross-links are set between the proteins forming a network matrix. The injection of the plasticized (propylene glycol) helps maintain the desired humidity level for storage. In order to dissolve and simultaneously activate the enzyme, milk whey was used. It is a by-product of milk processing containing milk plasma proteins, vitamins, minerals and traces of fats [17]. Milk whey proteins have high affinity with transglutaminase [18] so that its presence in the composition improves the physical and mechanical properties of the films produced. Besides, the prime cost of the finished film is reduced 2 times due to the use of cheaper milk whey [19].

The composition of the biodegradable film is shown in Table 2.

| No. | Components | %, mass |
|-----|------------|---------|
| 1.  | Propylene glycol | 12.5 – 14.6 |
| 2.  | Gelatin, 15%, water solution | 21.0 – 25.4 |
| 3.  | Transglutaminase, mixed with milk whey at the rate of | 42.5 – 51.0 |
|     | 1:5        |         |

The film is formed at room temperature and standard atmospheric pressure in 20-22 hours. The film produced is elastic, does not crack when bent, recovers its original shape easily when unbent, can take virtually any shape, has no smell and a neutral taste. The authors tried to assess the degradation of films in the natural environment. The freshly-made film degraded in six days. After three months of storage, one of the films was placed in the same conditions and it degraded in 20 days. The third film, stored for six months, degraded in 28 days [20, 21]. The durability and elasticity of the produced film were also tested.

5. Conclusion
Thus, the produced film is biodegradable and can be used for the production of biodegradable packaging materials, including for the food industry.

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