

## RECYCLING OF MULTILAYER POLYMER PACKAGING DOYPACK

## УТИЛІЗАЦІЯ БАГАТОШАРОВОЇ ПОЛІМЕРНОЇ УПАКОВКИ ДОЙ-ПАК

**Purpose of work.** The purpose of this work is to study the range of polymer multilayer composite packaging doypack in retail chains and finding possible ways to solve the problem of its recycling.

**Methodology.** Research on the range of polymer multilayer doypack packaging in Dnipro retail chains was conducted by selecting 20 different groups of goods in the three largest retail chains in Dnipro – ATB, Silpo and Varus. The composition of the combined package was determined by studying the appropriate labelling. Investigations of the properties of the crushed polymer multilayer doypack packaging were carried out using a microscope.

**Scientific novelty.** It was found that the combined polymer packaging doypack, used in retail chains in Dnipro, mainly consists of combined polyethylene (55-60%), combined polypropylene (20%), combined paper (5%), combined polypropylene with aluminium and polyethylene terephthalate (1% in the Silpo chain), combined polyethylene with polyethylene terephthalate (1% in the ATB chain), as well as those types of packaging where there is no labelling or is marked “7 OTHER” (14-19%). After shredding the doypack packaging into fractions smaller than 200 microns, part of the aluminium layer remains connected to the plastic part, which complicates further processing. The optimal temperature for the processing of the investigated waste by the extrusion method depends on the type of waste composition and for the polyethylene or polyethylene + polypropylene variant is 190-230 °C.

**Conclusions.** Recycling of polymer multilayer composite packaging is still a complex technological process, the success of which is influenced by many factors (incompatible polymers in different layers of the same film; adhesive additives that change the properties of polymers; non-plastic layers - aluminium and paper) and finally the composition of the package and its purity play a role in this. Because there is no system of collection and recycling of such packaging in our country, the bulk of plastic waste after use goes into the environment, and in particular – in aquatic ecosystems. There is a possibility that the polymer multilayer composite packaging doypack is a source of contamination of the aquatic environment with microplastics. The method of processing doypack polymer multilayer packaging, which includes shredding and subsequent extrusion or thermoforming is proposed. The solution to the problem could be expanded producer responsibility, as it encourages producers to consider the environmental impact at the stage of production, thus reducing waste and increasing the degree of packaging recycling through mandatory disposal and collection of used goods. The implementation of such a strategy in Ukraine would force the producers of combined polymer packaging to reconsider the technical characteristics of their products and make it more suitable for recycling.

**Keywords:** waste, multi-layered combined doypack packaging, recycling, environmental pollution, extended producer responsibility.

**Relevance of the problem.** People have used various packaging for goods, supplies, etc. almost throughout history. These were usually materials of natural origin

(leather, paper, plant leaves, banana leaves, various fabrics). Such materials were biodegradable, which is one of their advantages. Since the beginning of the scientific and technical revolution of the XIX-XX centuries and so far, numerous new types of materials - polymers - have been invented and synthesized. They were synthesized from fossil fuels. Subsequently, with the increase in the production of such materials, they began to be used for packaging in the food, cosmetic and other industries. The advantages of such packaging are that it is cheap, products can be stored in it much longer than in similar packaging, for example, made of paper; it is unattractive to animals and insects, it can be produced not only from 100% plastic, but also by combining it with paper or aluminium foil [1].

Polymeric materials effectively protect products from microbial damage and the influence of harmful environmental factors (sunlight, increased temperature and humidity, oxygen in the air, mechanical and chemical pollution, etc.), which increases the shelf life of products and reduces the amount of waste due to spoilage, especially during transportation and sales.

This study is about the type of packaging «doypack» or polymer multilayer combined packaging. It is a plastic bag with a bottom, which in a vertical position allows you to stand in a filled form. The main feature of the doypack is a multi-layer (from 1 to 9 layers) design with a flexible bottom. Packaging layers include various types of polymers, sometimes paper and aluminium foil. The packaging got its name in honour of the inventor – Louis Doyen [2].

Doypack packaging is used for packing coffee, tea, animal feed, sauces, nuts, crackers, chips, some baby juices, creams, toothpastes, chewing gums, etc.

Along with the many advantages of such packaging, there is also a serious problem - because the combined multilayer packaging contains several materials in its structure, it is practically not recyclable due to the complexity and high cost of the corresponding technologies. As a result, tons of this packaging end up in landfills every year, creating a number of environmental problems.

**Analysis of recent research and publications.** In modern literature, there are certain proposals for recycling such packaging. Thus, there is a technological scheme of a complex technological process of recycling waste from the production and consumption of composite layered aluminized packaging materials [3]. Waste from the packaging production or consumption is sent to a crusher for grinding, after grinding, polyethylene and aluminium foil are fed by a screw feeder into a reactor with a stirrer, which continuously receives a solution of acetic acid for chemical separation of polyethylene and aluminium layers. The author of the work noted that the complexity of such a processing scheme causes a low probability of trouble-free operation of the equipment. The main problems relate to two areas: with conveyors unloading raw materials, as well as with a press for forming products.

Unilever has patented the CreaSolv® multi-layer polymer packaging processing technology, developed to the company's order by the Fraunhofer Institute for Process Engineering and Packaging IVV. This solution has been successfully implemented and is working in Indonesia [4]. Due to its energy balance, this process has proven itself very well in environmental impact assessment studies [5].

British baby food brand Piccolo has announced the launch of the first 100% recyclable baby food pouch. Instead of producing a doypack with several layers of different materials, Piccolo offers a package consisting of only one layer of polypropylene with a lid and spout made of low-pressure polyethylene [6].

The previously widespread method of incineration of polymer waste finds more and more opponents, as it requires large costs for capturing gaseous toxic products. Pyrolysis, which is a high-temperature decomposition of waste without access to oxygen with the production of semi-coke and combustible gas, is currently considered as one of the alternative ways of processing polymer waste, but it also has disadvantages. Chemical destruction methods and biological methods are considered promising, but today they are limited in application due to sufficiently high requirements for molecular homogeneity and purity [7].

Therefore, the analysis of the latest research and publications showed that the processing of polymer multilayer combined doypack packaging is a complex technological process, the success of which is influenced by numerous factors (incompatible polymers in different layers of the same film; adhesive additives that change the properties of polymers; non-plastic layers – aluminium and paper) and not the least role in this is played by the composition of the packaging and its cleanliness.

**Purpose of work.** Therefore, the purpose of this work is to study the range of polymer multilayer composite packaging doypack in retail chains and finding possible ways to solve the problem of its recycling.

**Presentation of the main research material.** The research was conducted in the three largest supermarket chains in the city of Dnipro. These are the «ATB», «Silpo» and «Varus» chains.

It should be noted that some of the goods on the shelves do not have exact markings, but according to their appearance and physical properties, they are packed in such a package. Among them are already listed groups of products (coffee, tea, animal feed, sauces, nuts, crackers, chips, some baby juices, creams, toothpastes, etc.), as well as, for example, meat and sausage cuts.

20 different products from three store chains were selected for analysis. The period of the experiment is October 2021. The results of the research are shown in figures 1-3.

As we can see from the data shown in the figures, the most common types of doypack packaging are: combined polyethylene (55-60% of all packaging), combined polypropylene (20%), combined paper (5%), combined polypropylene with aluminium and polyethylene terephthalate (1% in the «Silpo» network), combined polyethylene with polyethylene terephthalate (1% in the «ATB» network), as well as types of packaging where there is no marking or there is a «7 OTHER» mark (14-19%).

Also of particular concern is the significant percentage of packaging marked «7 OTHER» (14-19%) – because this type of packaging is not recyclable at all and the expediency of producing such packaging from an ecological point of view is extremely questionable.

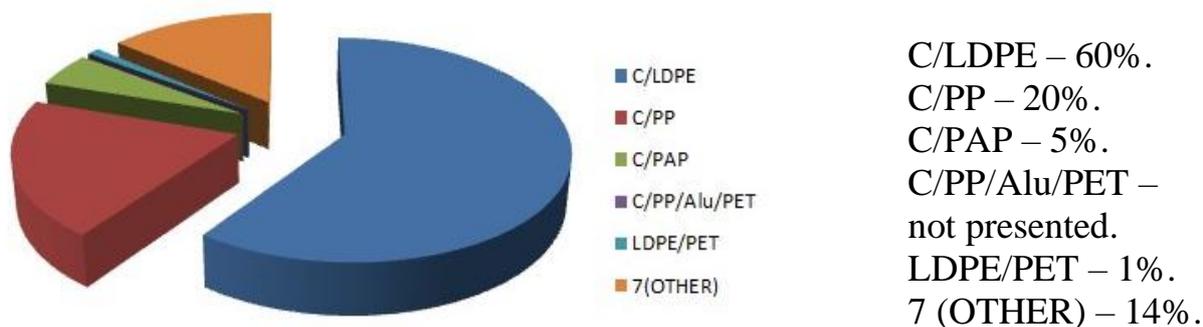


Fig. 1. The results of research on the assortment of doypack packaging in the «ATB» supermarket chain

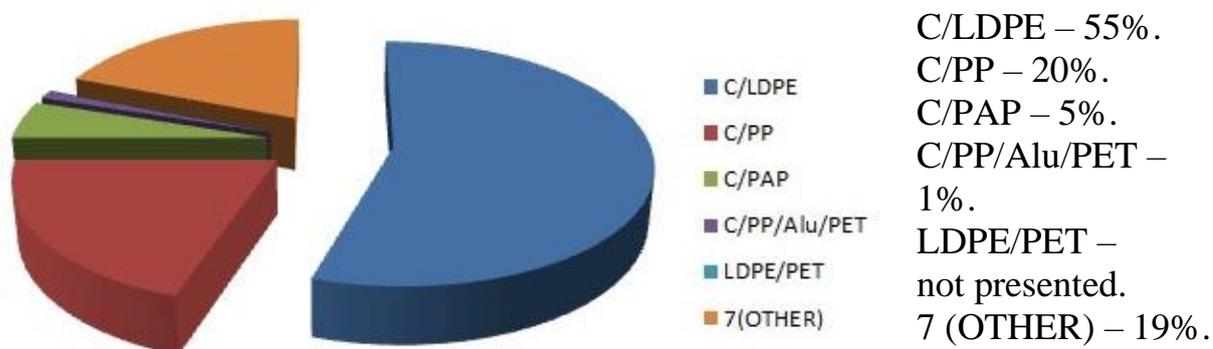


Fig. 2. The results of research on the assortment of doypack packaging in the «Silpo» supermarket chain

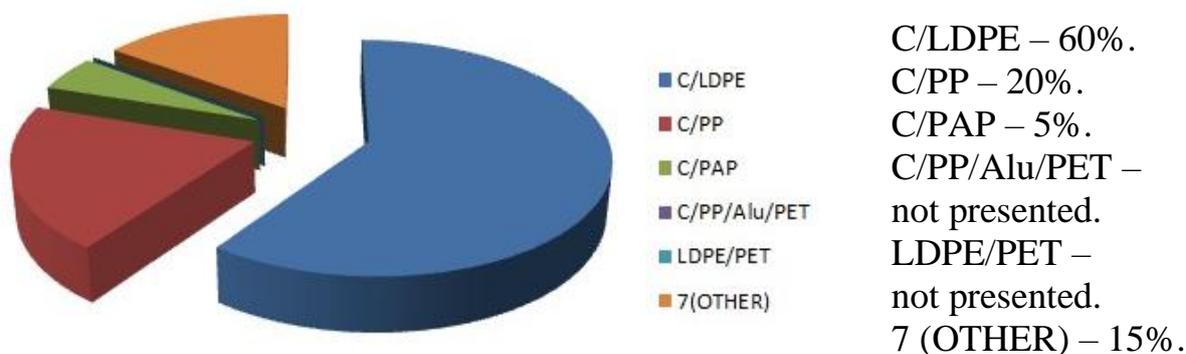


Fig. 3. The results of research on the assortment of doypack packaging in the «Varus» supermarket chain

The main manufacturers of doypack packaging that took part in this study in Ukraine are the following companies: «HTM Limited Ukraine» (Kyiv), «TERA PLAST» (Odesa), «NOVOPLAST» (Odesa), LLC MNVP «Aris» Ltd. (Kharkiv), «STARFLEX» (Kharkiv), «TM PAKADO» (Vyshhorod) and others.

Due to the wide variety of possible combinations of polymers, it is very difficult to study them in detail. Accordingly, it is an almost impossible task to divide the films

among themselves by type. In addition, the experience of processing waste of combined films made of heterogeneous polymer materials shows that the obtained regranulate is very difficult to use to produce new products [8].

As a result of the fact that in our country there is no system of collection and secondary processing of such packaging, the main part of plastic waste after use ends up in the environment, and in particular – in water ecosystems. At the same time, the process of complete decomposition of plastic in natural conditions can take hundreds and thousands of years.

In addition to soil and air pollution, plastic fragments entering the aquatic environment are of particular concern. While in water, plastic products gradually break down due to exposure to sunlight, oxidation or physical wave action and currents, because of which a huge number of small particles are formed, which have recently been called «microplastics».

Pollution of the aquatic ecosystems with microplastics is dangerous for the environment and especially for human health, however, the nature of possible negative consequences has not yet been determined and the maximum permissible concentration of microplastics has not been established.

Corresponding studies on the contamination of the aquatic environment with microplastics were conducted in the water area of the city of Dnipro [9]. Microplastic particles were registered in all studied water samples of the Dnipro River. The concentration of microplastics ranged from 0.4 to 10 pcs. / dm<sup>3</sup>, the average concentration of microplastics in surface river water within the central part of the city of Dnipro was 3.4 pcs. / dm<sup>3</sup>. Both primary microplastics (foam balls) and secondary microplastics were found in the analysed samples. This indicates that the storm drain that passes under the city centre is a serious source of plastic pollution of the river water, and it cannot be ruled out that, in addition to microparticles of plastic bottles, plastic bags, disposable tableware and others, the river water contains decay products of doypack packaging.

The next purpose of the study was to find ways to recycle polymer multi-layer doypack packaging. For the research, production waste and exclusively clean, sorted doypack packaging from various products were used. Shredding of doypack packaging into several different fractions was carried out. The size of each fraction was +1000, +1000–500, +500–200 and smaller than 200 µm. The appearance of several fractions is shown in the Fig. 4.

The inlet temperature before shredding is + 20 °C. The mass of the assembled packaging for the experiment is 25 kg. At the exit, the temperature of the raw material was +60 °C for all fractions of the material. The crusher was made by our own hands, a 2-kW electric motor was used. The productivity of this crusher is several tens of kilograms of crushed material per hour, depending on the input fraction of the material. It should be noted that depending on the incoming fraction of the material, the power of the crusher must be calculated so that it can cope with its task, and so that large packaging does not lead to clogging of the receiving hopper of the crusher and failure of the mechanical part or electric motor due to overloads during operation. Plastic shredding is a good and technologically simple method for recycling plastic. Some

types of secondary raw materials can only be crushed, which is necessary for their future processing and production of other products from secondary plastic.

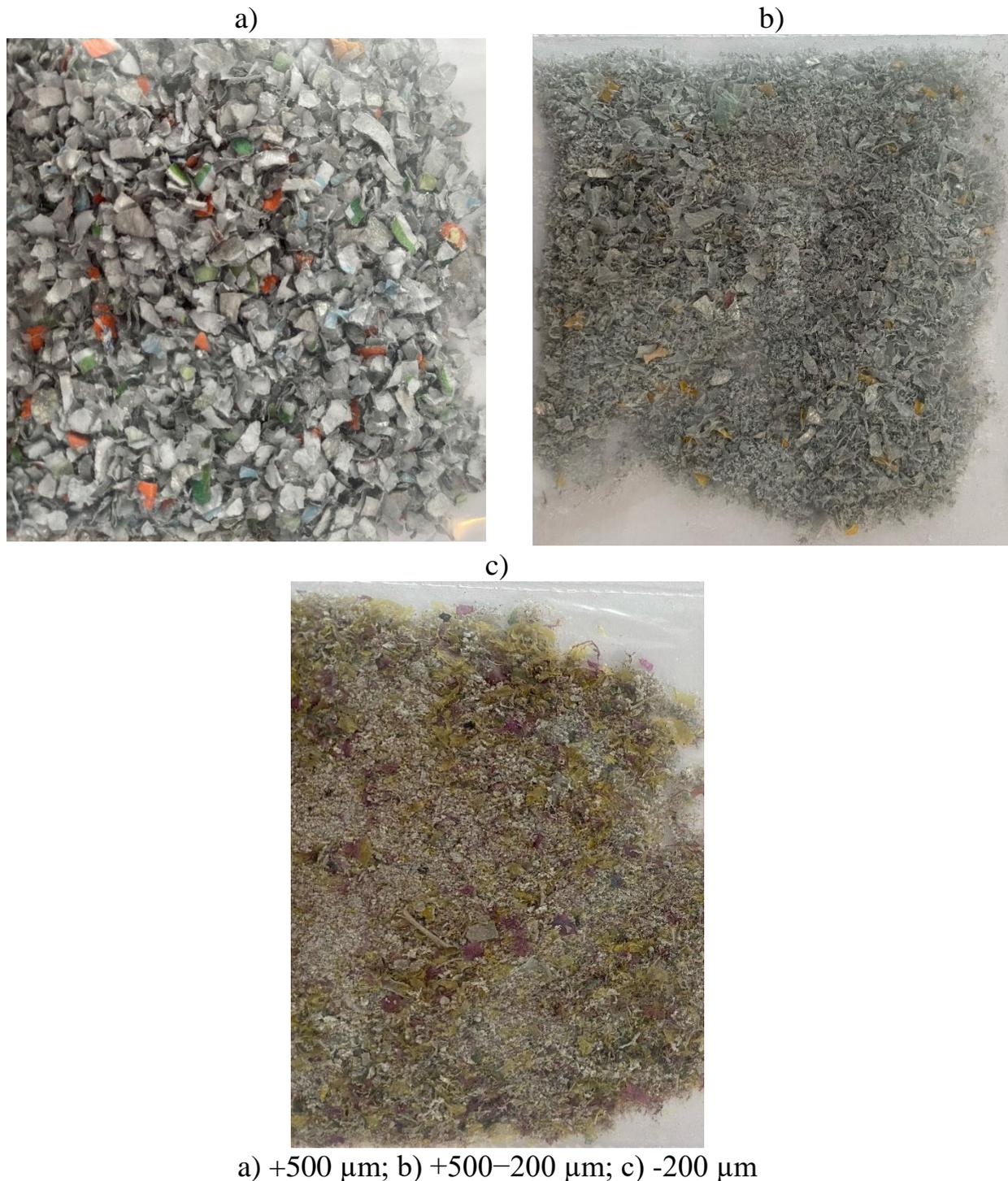


Fig. 4. Fractions of crushed polymer multi-layer packaging doypack

An analysis of the qualities of the crushed polymer multilayer doypack packaging was also carried out using a microscope. It was found that after crushing the doypack packaging, part of the aluminium layer remains connected to the plastic part, which complicates further processing. If during processing there is no purpose to separate aluminium from plastic, then after crushing, such material can be reused for secondary

moulding from plastic, as well as raw material for extrusion and manufacturing of various materials (for construction, etc.). A photo of a shredded polymer multilayer doypack package from a microscopic study is shown in the Fig.5.

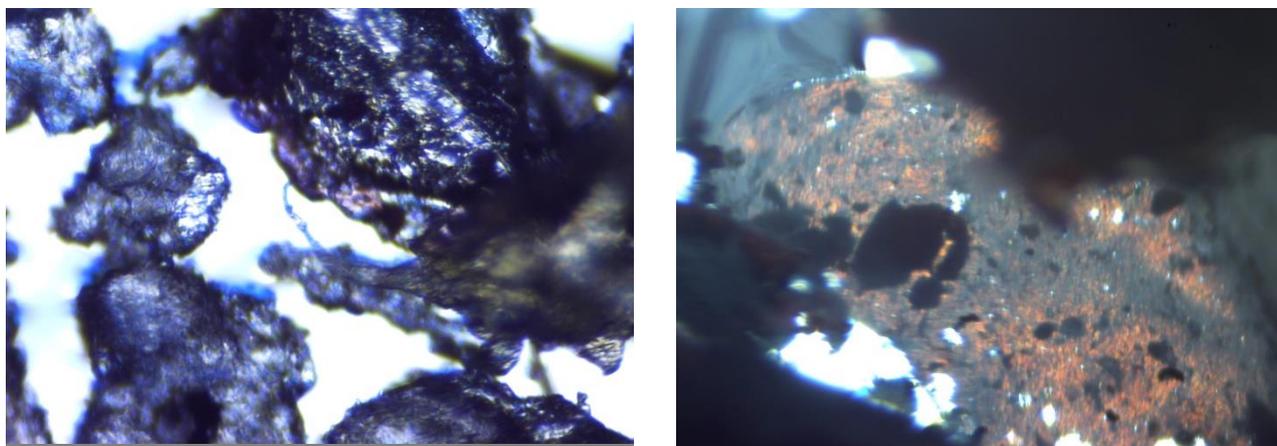


Fig. 5. Shredded doypack (magnification 200x)

The next stage of processing was the extrusion of this material. Extrusion is a type of technological process during which the material is moved with the help of a screw inside the «sleeve» of the extruder body, where a viscous and homogeneous mass of material is formed under the influence of pressure and temperature. The extruder for the experiment was made by our own hands, the main electric motor had a power of 1.5 kW, as well as 4 heating elements with a total power of 1 kW. After the mass exits the auger, it can be moulded into various products or, with the help of a special system for cutting and cooling the mass, obtain a secondary polymer granule. Extrusion was carried out at a temperature of +200 °C. In our case, we made a special mould with the help of which it is possible to obtain a square plastic beam at the output. It is worth noting that when processing plastic, which will include the remains of aluminium foil, it will be necessary to clean the filter nets on the sliders of the extruder screw more often, as well as replace the metal knives more often, if this type of processing material will be used to produce secondary polymer granules. The plastic beam obtained because of extrusion is shown in Fig. 6.



Fig. 6. Plastic beam 34/34 mm, made of doypack

It is worth noting that to obtain a dense and strong material, it is necessary to monitor the uniform supply of raw materials to the «sleeve» of the extruder, as well as to choose the right temperature for the type of polymer that is most often used in doypack packaging. If it will be polyethylene, or for example polyethylene + polypropylene, then the optimal temperatures for extrusion will be +190-230 °C. The more correctly the melting temperatures are set during processing, the more times the material can be recycled. This is because at temperatures higher than recommended, the chemical structure of the polymer is destroyed. It is also desirable that the material was washed from various organic residues (so that during extrusion the material does not start burning inside the «sleeve» of the extruder and there are no voids in the obtained material from combustion gases of organic residues).

If we heat the doypack crushed on a crusher without using an extruder, where pressure is created during the operation of the screw inside the “sleeve”, and for this we use, for example, an electric oven and heat the crushed material in a special metal form at + 200 °C, we can also get material from secondary plastic, which can be used both for production and decoration. The disadvantage of this material is less strength, it has many pores in its structure, it is much easier to break it. An example of thermoforming is given below, in Fig. 7.



Fig. 7. The obtained material after thermoforming

It should also be noted that, in general, the problem of waste recycling should be solved by the producers of this waste, and not by the final consumers. Many countries have long implemented national packaging waste management systems with positive results. The basis of these systems are the Directives of the European Parliament and the Council 94/62/EC and 2004/12/EC “On packaging and packaging waste”. In these systems, the principle of extended producer responsibility applies – this is an approach within the framework of environmental policy, according to which the responsibility of the producer for the product extends to the post-sale stage of its life cycle. That is, it is the manufacturers of the products, not the users, who are responsible for accepting the returned products and the waste left after the use of such products, as well as the subsequent management of the waste and the financial responsibility for such activities [10].

Such a management strategy already at the stage of production prompts manufacturers to consider environmental consequences, thanks to which the generation of waste is reduced and the degree of recycling of packaging is increased due to the mandatory disposal of released goods and the mandatory collection of used goods. The implementation of such a strategy in Ukraine would also force manufacturers of doypack combined polymer packaging to review the technical characteristics of their products and make them more suitable for recycling.

### **Conclusions and prospects for further research.**

1. The processing of polymer multilayer combined doypack packaging is a complex technological process, the success of which is influenced by many factors (incompatible polymers in different layers of the same film; adhesive additives that change the properties of polymers; non-plastic layers – aluminium and paper). The composition of the packaging and its cleanliness play an important role in this matter.

2. The most common types of doypack packaging on the shelves of large retail chains of the country are: combined polyethylene (55-60%), combined polypropylene (20%), combined paper (5%), combined polypropylene with aluminium and polyethylene terephthalate (1% in the "Silpo" chain), combined polyethylene with polyethylene terephthalate (1% in the "ATB" chain), as well as types of packaging where there is no marking or there is a "7 OTHER" mark (14-19%).

3. As a result of the fact that in our country there is no system of collection and recycling of such packaging, the main part of plastic waste after use ends up in the environment, and in particular – in water ecosystems. There is a possibility that the polymer multi-layer combined doypack packaging is a source of contamination of the aquatic environment with microplastics.

4. The possibility of processing the combined polymer packaging doypack by grinding and subsequent extrusion or thermoforming was investigated. To obtain a high-quality processing product, it is extremely important to observe a certain temperature regime. This will allow you to repeat the recycling process several times.

5. The extended responsibility of the manufacturer already at the stage of production encourages manufacturers to consider the environmental consequences, thanks to which the generation of waste is reduced and the degree of recycling of packaging is increased. The implementation of such a strategy in Ukraine would force manufacturers of doypack combined polymer packaging to review the technical characteristics of their products and make them more suitable for recycling.

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#### АНОТАЦІЯ

**Мета.** Метою даної роботи є дослідження асортименту полімерної багатошарової комбінованої упаковки дой-пак у торговельних мережах та пошук можливих шляхів вирішення проблеми її утилізації.

**Методологія.** Дослідження асортименту полімерної багатошарової упаковки дой-пак у торговельних мережах м. Дніпро проводилися шляхом відбору 20 різних груп товарів у трьох найбільших торговельних мережах м. Дніпро – АТБ, Сільпо та Варус. Склад комбінованої упаковки встановлювався шляхом вивчення відповідного маркування. Дослідження властивостей подрібненої полімерної багатошарової упаковки дой-пак виконувались за допомогою мікроскопу.

**Наукова новизна.** Встановлено, що комбінована полімерна упаковка дой-пак, що використовується у торговельних мережах м. Дніпро, в основному складається з комбінованого поліетилену (55-60%), комбінованого поліпропілену (20%), комбінованого паперу (5%), комбінованого поліпропілену з алюмінієм та поліетилентерефталатом (1% в мережі «Сільпо»), комбінованого поліетилену з поліетилентерефталатом (1% в мережі «АТБ»), а також таких видів пакування, де відсутнє маркування або є позначка «7 OTHER» (14-19%). Після дроблення упаковки дой-пак до фракцій розміром менше 200 мкм частина алюмінієвого шару залишається з'єднаною з пластиковою частиною, що ускладнює подальшу переробку. Оптимальна температура для переробки досліджуваних відходів методом екструзії залежить від складу відходів і для варіанту поліетилен або поліетилен + поліпропілен становить +190–230 °С.

**Висновки.** Переробка полімерної багатошарової комбінованої упаковки дой-пак є складним технологічним процесом, на успішність якого впливає велика кількість факторів (несумісні між собою полімери в різних шарах однієї плівки; адгезивні добавки, що змінюють властивості полімерів; непластикові шари – алюміній та папір) і не останню роль у цьому відіграє склад упаковки та її чистота. Внаслідок того, що в нашій країні відсутня система збору та вторинної переробки подібного пакування, основна частина пластикових відходів після використання потрапляє в навколишнє середовище, і зокрема – в водні екосистеми. Існує ймовірність того, що полімерна багатошарова комбінована упаковка дой-пак є джерелом забруднення водного середовища мікропластиком. Запропонований метод переробки полімерної багатошарової упаковки дой-пак, що включає подрібнення та подальшу екструзію або термоформування. Рішенням проблеми могла би стати розширена відповідальність виробника, адже ця система вже

на етапі виробництва продукції спонукає виробників враховувати екологічні наслідки, завдяки чому скорочується утворення відходів і підвищується ступінь переробки упаковки за рахунок обов'язкової утилізації випущених товарів та обов'язкового збору використаних товарів. Впровадження такої стратегії в Україні змусило би виробників комбінованої полімерної упаковки дій-пак переглянути технічні характеристики своєї продукції і зробити її більш придатною для вторинної переробки.

**Ключові слова:** *відходи, багат шарова комбінована упаковка дій-пак, вторинна переробка, забруднення довкілля, розширена відповідальність виробника.*