Effect of PP+EVOH regranulate core thickness on mechanical properties of co-injection molded multilayer thin-wall packaging

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Abstract: The co-injection process of thin-walled, three-layer packaging with a core of regranulate r(PP+EVOH) was investigated. Increasing the core content (10–50 v/v%) resulted in a reduction in the thickness of the outer PP layers, tensile strength (21%), Young modulus (17%), and compressive force (9%). The molded parts retained properties suitable for packaging applications. The feasibility of incorporating up to 50 v/v% of r(PP+EVOH) into the core was confirmed.

Keywords: co-injection molding, thin-walled packaging, mechanical properties, mechanical recycling, circular economy.

Wpływ grubości rdzenia z regranulatu PP+EVOH na właściwości mechaniczne współwtryskiwanych wielowarstwowych opakowań cienkościennych

Streszczenie: Zbadano proces współwtryskiwania cienkościennych, trójwarstwowych opakowań z rdzeniem z regranulatu r(PP+EVOH). Zwiększenie udziału rdzenia (10–50% obj.) powodowało zmniejszenie grubości warstw zewnętrznych z PP, wytrzymałości na rozciąganie (21%), modułu sprężystości (17%) oraz siły ściskającej (9%). Wypraski zachowały właściwości odpowiednie do zastosowań w branży opakowaniowej. Potwierdzono możliwość wprowadzenia do 50% obj. regranulatu r(PP+EVOH) do rdzenia.

Słowa kluczowe: współwtryskiwanie, opakowania cienkościenne, właściwości mechaniczne, recykling mechaniczny, GOZ.

Since the 1950s, global plastic production has grown quickly, reaching a record 4.2×10^{11} kg in 2023 [1]. This growth is the result of the wide use of plastics in many industries, thanks to their low weight, high strength, easy processing, and relatively low production cost [2]. However, most plastics do not break down naturally, and many products are used for only a short time. This leads to a steady increase in plastic waste. In 2018, the total global amount of plastic waste was more than 3.4×10^{11} kg, with packaging as the largest source, followed by construction and automotive industries [3]. More than half of all plastic waste comes from single-use products with a life of less than one month [5].

Even though recycling rates are improving, most food packaging made from polyolefins such as polypropylene (PP) and polyethylene (PE), as well as polyethylene terephthalate (PET), is not recycled. About 90 percent of such packaging is sent to landfills or burned, which causes annual economic losses of 80 to 120 billion US dollars [6]. Recycling is difficult because many packages are made in multilayers form with the use of different plastics, which are hard to separate and identify. Contamination from food and the content of additives such as colors, stabilizers, and fillers also reduce the quality of recycled materials [7].

Today's packaging design must meet high demands for strength, barrier properties, and product protection, while also supporting environmental goals. Designers now consider the whole life cycle of packaging, including recycling and lowering carbon emissions. Modern solutions combine protective functions with convenience features such as resealable closures, easy opening, and smart labels. The choice of production method depends on the type of plastic, the intended use of the packaging, storage conditions, and required mechanical and functional properties. Food packaging is made using methods such as injection molding [8], extrusion [9], and thermoforming

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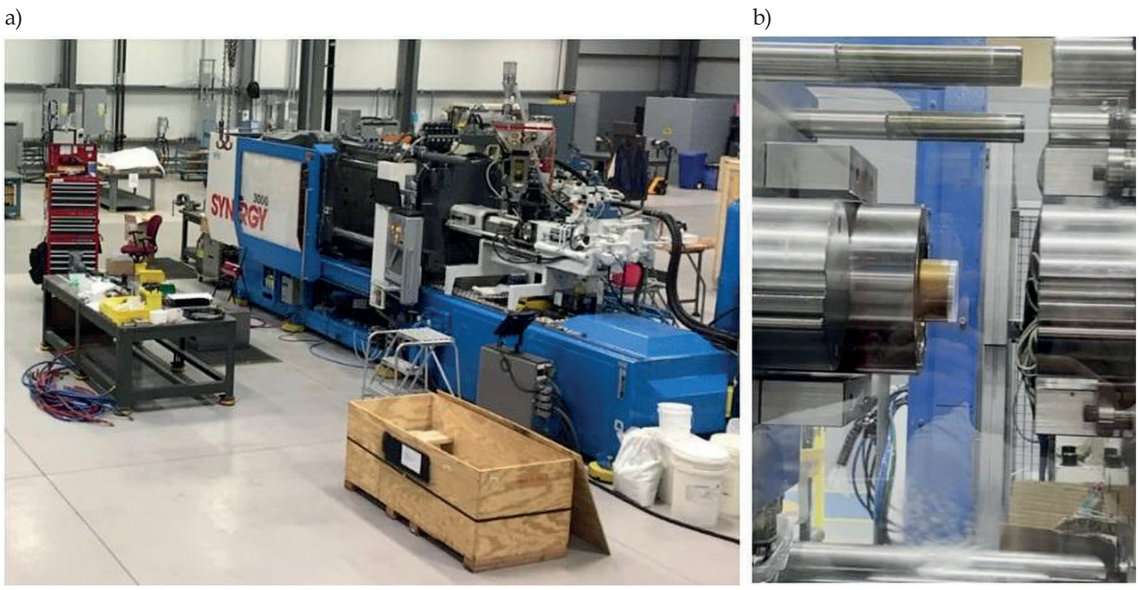

Fig. 1. Research station: a) injection molding station, b) injection mold with a hot runner system for COI technology

[10], as well as advanced techniques like co-extrusion [11] and co-injection molding [12]. These advanced methods make it possible to produce multilayer packaging with specific barrier properties and gas permeability, helping to keep food fresh and maintain its quality.

Co-injection molding (COI) is one of the most effective techniques for producing multilayer packaging in a single production cycle. It uses two different thermoplastics to form outer and inner layers with distinct technical and functional roles [14, 15]. This method supports one of the leading trends in polymer engineering, which is to give products additional useful properties while enabling the incorporation of post-consumer recycled (PCR) mate-

T a b l e 1. Co-injection molding parameters

Parameter	r(PP+EVOH) content, v/v%		
	10	30	50
Temperature PP, °C	240	240	240
Temperature r(PP + EVOH), °C	240	240	240
Injection flow length PP, mm	39.47	29.34	22.12
Injection flow length r(PP + EVOH), mm	21.41	45.06	51.97
Injection time PP, s	0.72	0.74	0.70
Injection time r(PP + EVOH), s	0.54	0.22	0.20
Injection delay r(PP + EVOH), s	0.06	0.45	0.45

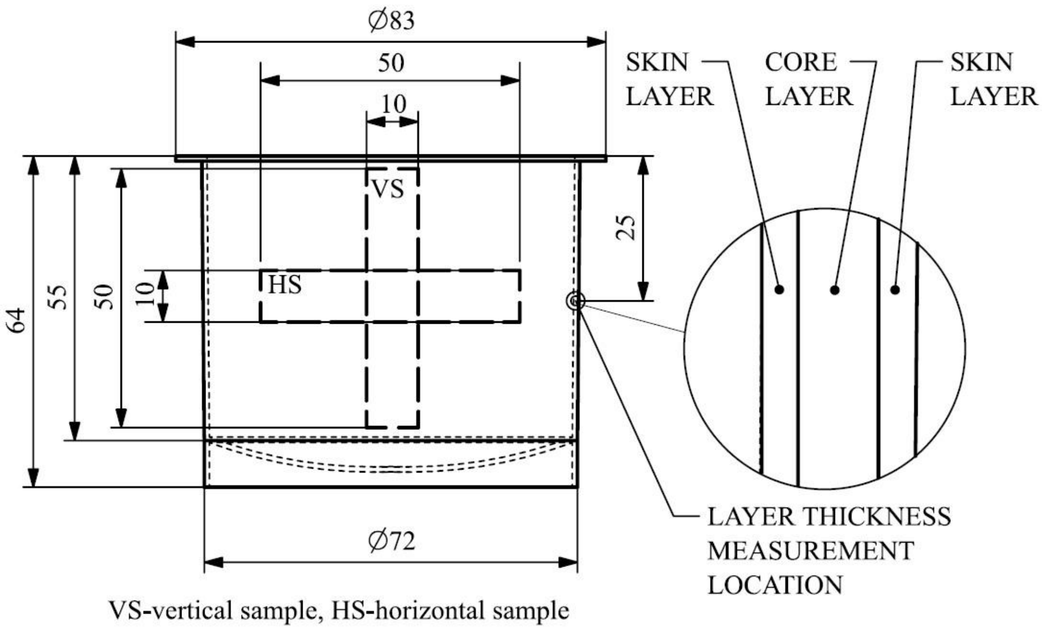

Fig. 2. Characteristics of geometric features and sampling locations for mechanical properties testing

rials in selected layers. European Commission guidelines within the Circular Economy Action Plan not only permit but, in some product categories, actively encourage the use of PCR in functional layers of new packaging [16]. The correct choice of material combinations is essential to ensure interfacial compatibility, good processing performance, and high product quality.

The aim of the paper is to assess the possibility of using co-injection molding technology to produce thin-walled, three-layer packaging with a core layer made of PP+EVOH blend recycled from barrier packaging waste. The study focused on evaluating the distribution of layer thickness in the walls of thin-walled packaging. The aim was to obtain the maximum volume of recycled PP+EVOH blend that could be introduced into the core. Additionally, the effects of molecular orientation and shear stresses on the mechanical properties of 3-layer thin-walled moldings with a high recycled blend content were investigated.

EXPERIMENTAL PART

Materials

Polypropylene (PP) Moplen RP390T from LyondellBasell (Rotterdam, Netherlands) was used as the skin layer material, while the core layer consisted of a recycled blend of PP (Moplen RP390T) and ethylene vinyl alcohol (EVOH) XEP-1248 from EVAL (Okayama, Japan). Melt flow rate (MFR) for used PP was 40 g/10 min and 18 g/10 min for EVOH.

Samples preparation

Three-layer co-injection molded parts with an approximate wall thickness of 1 mm were produced with a core layer containing 10, 30, and 50 v/v% recycled PP+EVOH blend. The process was performed on a Netstal S 3000-230/60 two-component injection molding machine (Näfels, Switzerland) with a pressing force of 3000 kN, equipped with screws with a diameter of 38 mm for the primary component (shell) and 18 mm for the secondary component (core). A specialized single-cavity injection mold with a co-injection molding (COI) hot runner system from Mold-Masters (Georgetown, Canada) was used. The hot runner system included needle valves controlled by pneumatic actuators. Fig. 1 shows a production cell equipped with a Netstal injection molding machine and an injection mold with a special hot runner system for co-injection molding.

Most essential co-injection molding parameters are shown in Table 1.

The regranulate r(PP+EVOH) was produced from 3-layer PP/EVOH/PP packaging that were reground on modified, original research stand described in article [17]. A twin-screw extruder Leistritz ZSE18 Maxx (Nuremberg, Germany) was used to prepare the regranulates.

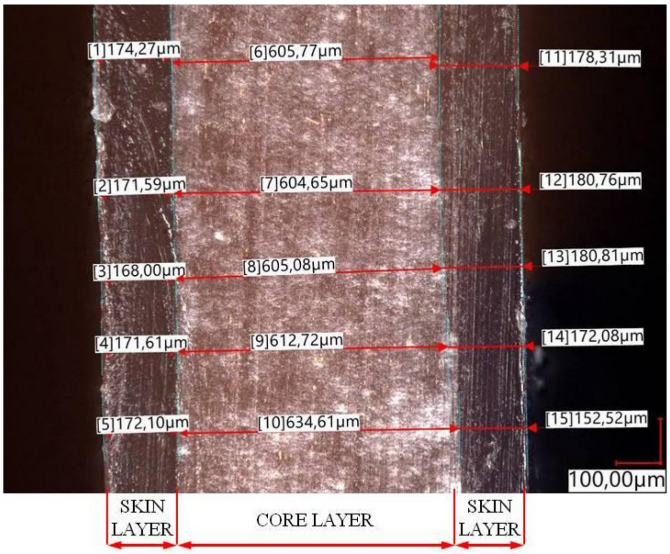

Fig. 3. Example of measuring the thickness of the outer PP layers and the core r(PP+EVOH) layer

Tensile tests were conducted on specimens measuring 50×10×1 mm. Samples were cut horizontally and vertically from the side walls of the packaging. This approach enabled the assessment of tensile behavior parallel and perpendicular to the melt flow direction in the mold cavity, allowing evaluation of the influence of macromolecular orientation on the resulting mechanical properties. The sampling location is shown on Fig. 2.

Methods

Both compression and tensile tests were performed on a Zwick/Roell Z030 universal testing machine (Ulm, Germany), at a crosshead speed of 50 mm/min and gauge length of 50 mm. Young's modulus was evaluated at a crosshead speed of 1 mm/min. Tensile tests were performed in accordance with the PN-EN ISO 527 standard; however, the samples did not meet the standard's requirements due to dimensional limitations of the primary test object.

The compression tests were carried out with the samples placed "bottom up" on the compression plate at a compression speed of 10 mm/min, in accordance with ISO 12048.

Table 2. Tensile properties of the samples

r(PP+EVOH) v/v%	Cut vertically to the axis		Cut horizontally to the axis	
	Young modulus MPa	Tensile strength MPa	Young modulus MPa	Tensile strength MPa
10	741±55	30.3±0.3	662±22	27.2±0.8
30	620±37	25.8±0.3	589±11	24.1±0.7
50	616±49	24.0±0.3	554±6	22.5±0.1

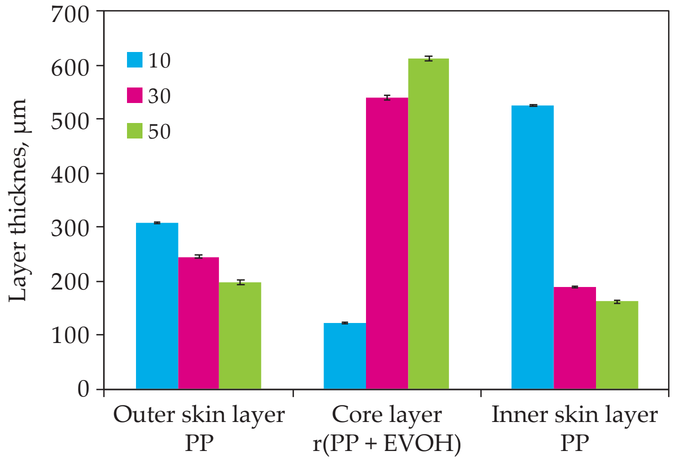

Fig. 4. Influence of r(PP+EVOH) content on the thickness of the layers

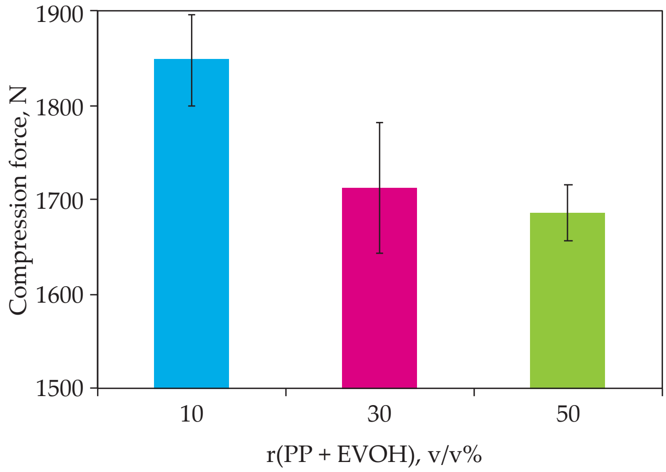

Fig. 5. Influence of r(PP + EVOH) content on the maximum compressive force

The thickness of each layer in the multilayer structure was measured using a Keyence VHX-7000 digital optical microscope (Osaka, Japan) equipped with a VH-Z100R lens. The analysis was performed using composite spatial imaging by capturing a series of images at different focal planes, ranging from the highest to the lowest points of the cross-sectional surface. These images were then automatically combined into a single sharp image using the depth composition function. Images were taken at 100 × magnification with ring lighting, and a depth composition step of 15 µm was applied. Test samples were previously cut longitudinally to expose a full cross-section of the samples. Thickness measurements were taken 25 mm from the flange of the packaging. Five independent measurements were performed for each of the three layers: outer (PP), middle r(PP/EVOH), and inner (PP). The arithmetic mean and standard deviation were calculated from the obtained data.

RESULTS AND DISCUSSION

Co-injection molding allows the production of three-layer thin-walled moldings using regranulates as the core. Changing the core thickness while maintaining a constant wall thickness is effectively achieved by varying the injection delay time of the mechanically recycled material. Microscopic images of the three-layer wall cross-sections show that both materials, PP and

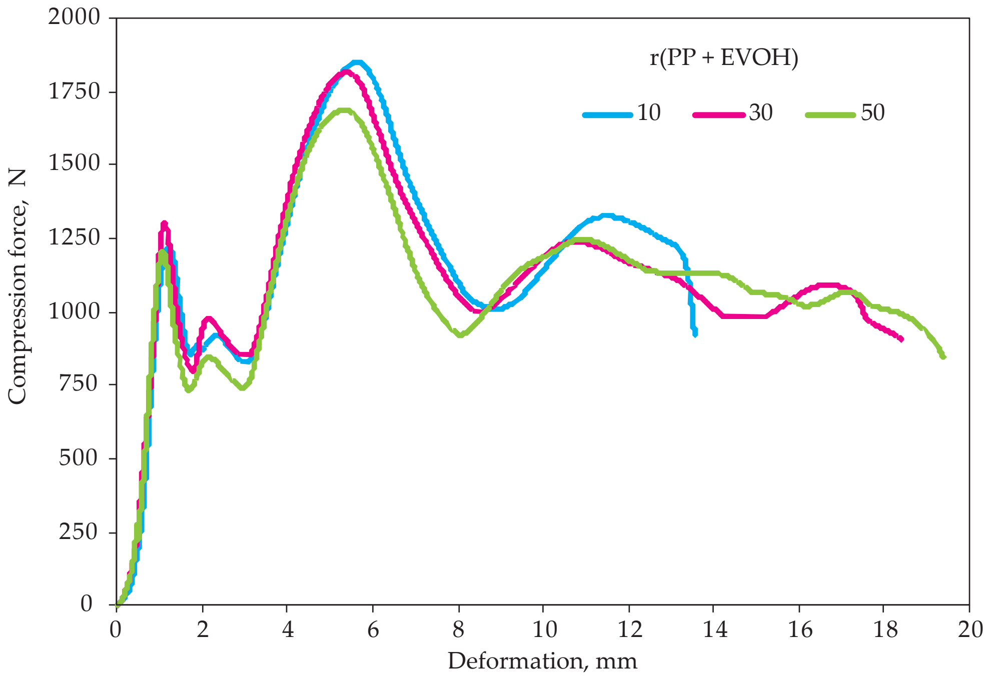

Fig. 6. Compressive force-deformation curves depending on r(PP+EVOH) content

r(PP+EVOH), flow laminarly to fill the injection mold cavity, maintaining the thickness relationships within the molded part along the entire wall length (Fig. 3). Within the tested range of regranulate content in the molded part core, a constant relationship was achieved between the thickness of the outer and inner layers relative to the core thickness. For molded parts with the highest regranulate content (50 v/v%), the outer PP layers had an average thickness of 171.51±2.26 μm, while the inner layer had an average thickness of 172.89±11.93 μm. A similar proportion of both PP layer thicknesses in the wall cross-section was observed, meaning that both layers were almost symmetrically positioned relative to the r(PP+EVOH) core. It was an essential improvement compared to variants with a lower content of reused material, which showed significant differences in thickness between the PP layers. The proportion of the core layer in the part's wall cross-section was observed to have a strong impact on its forming method. The phenomena described are consistent with the observations of Vangosa [18], who showed that increasing the volume fraction of the core material, regardless of the method of its introduction, leads to a reduction in the thickness of the skin layers and an increased degree of core penetration.

The analysis of the effect of the regranulate content r(PP+EVOH) on the formation of layers in the molded part wall is summarized in Fig. 4. The dosing of the smallest amount of core material was proportionally represented in the dimensional relations of the PP/r(PP+EVOH) layers. The main reason for the observed changes is the significant difference in viscosity of both materials at low injection speeds (PP has twice the MFR) [19].

The increase in r(PP+EVOH) content in the molded part wall has significant effect on the mechanical properties of samples cut from the three-layer PP/r(PP+EVOH)/PP molded part wall, both in the horizontal and vertical directions (Table 2). The samples are characterized by strength and Young's modulus in the ranges obtained by other researchers [20].

It can be observed that with the increase in the volume fraction of the core material, both the tensile strength (21%) and Young's modulus (17%) are significantly reduced, regardless of the stretching direction. Due to the increasing proportion of elasticity EVOH in the blend, which exhibits a lack of adhesion to polypropylene. This phenomenon can be observed for both types of samples. Samples cut parallel to the packaging axis have better strength properties, which result from the alignment of the macromolecules in the flow direction of the materials in the melted state.

The maximum compressive force that causes deformation of three-layer PP/r(PP+EVOH)/PP packaging, depending on the core layer content, are presented in Fig. 5. Compression tests were conducted with the packaging bottom facing up. A significant effect of the r(PP+EVOH) core material content on the dimensional stiffness of the sample was observed. Increasing the core content from 10 to 50 v/v% resulted in a decrease in the maximum compressive force from 1848 to 1686 N (by 9%).

The observed stiffness differences between samples containing from 10 to 50 v/v% of regranulate r(PP+EVOH) result from the increasing share of EVOH which is a component with poor adhesion to polypropylene, which increases the deformability of the material in the core layer and entire packaging (Fig. 6).

CONCLUSIONS

Co-injection technology was successfully used to produce thin-walled packaging moldings with a structure of at least three layers, in which the core layer was made of r(PP+EVOH) from recycled barrier packaging waste. The resulting moldings were characterized by a uniform thickness distribution of the PP/r(PP+EVOH)/PP layer, despite significant differences in the apparent viscosity of the polymers used. It was found that the thick distribution of the individual layers strongly depends on the flow and solidification conditions of the molten polymer in the injection mold cavity, as well as on the proportion of the r(PP+EVOH) regranulate layer in the core of the three-layer component. It was demonstrated that up to 50% v/v of regranulate can be incorporated into the core layer. The molding produced under high process pressure demonstrated favorable mechanical properties even without a compatibilizer. It can also be assumed that the orientation of macromolecules in the flow direction and high shear stresses may be responsible for maintaining or only slightly reducing the barrier properties, especially in the case of the highest r(PP+EVOH) content in the core layer.

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Authors contribution

P.W. – conceptualization, writing-original draft, investigation, methodology; D.S. – conceptualization, supervisor, methodology; P.C. – visualization, validation.

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Conflict of interest

The authors declare no conflict of interest.

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