Abstract

The research presented in this dissertation focuses on the analysis of the structure and properties of newly developed 3-layer composite films produced using blow moulding technology supported by a precision gravimetric dispensing system. In order to assess the influence of the addition of regranulate and mineral filler on the properties of the films intended for automated form-fill-seal (FFS) packaging lines, a number of tests and studies were carried out including artificial ageing of the films, FTIR (Fourier Transform Infrared Spectroscopy), DSC (Differential Scanning Calorimetry), OOT (Oxidative Onset Temperature), XRD (X-ray Diffraction) and mechanical tests, such as tensile strength, tear resistance, puncture resistance and also the coefficient of friction. Microscopic observations of the cross-sectional area of the film and SEM (Scanning Electron Microscopy) analysis of the fractures of prepared film samples were conducted.

As part of the research, three-layer LDPE (Low-Density Polyethylene) films were produced with the proposed layer distribution: A- 20%, B-60%, C-20%, with varying amounts of RPE1 regranulate (selected for the study) and calcium carbonate in the middle layer of the film. The films were produced on a Labtech Engineering three-layer laboratory line. Eight film samples were prepared, including a reference film labelled EI1 made exclusively from virgin films, and seven films with a modified middle layer B, containing regranulate and calcium carbonate in specified proportions.

In order to determine the structure and properties of the modified films during and after exposure to varying atmospheric conditions, an artificial ageing process was carried out in a climatic chamber, simulating the long-term effects of atmospheric factors such as humidity, temperature, and UV radiation on the structure and mechanical properties of the films. The results showed that artificial ageing under the specified conditions (*alternating UV light and moisture cycles at 60±3°C*) did not negatively affect the condition of the tested films. Taking this into account, X-ray diffraction and mechanical strength tests of the films and seals made on the films were conducted before and after the artificial ageing process. The results of these studies were crucial for verifying the durability of the packaging and seals stored under varying atmospheric conditions over time. The first step in the research was to explore how ageing processes affect the film's structure. XRD tests were carried out for this purpose. The analyses confirmed that the addition of regranulate and calcium carbonate does not cause degradation of the film's crystalline structure. Furthermore, the films that were subjected to artificial ageing showed a slightly higher proportion of the crystalline phase compared to films before the ageing

process, likely due to what is known as secondary crystallisation. Based on these results, mechanical tests were carried out, including tensile tests of both the regular films and the films with seals, measured along and across the extrusion direction. These tests were conducted both before and after the artificial aging process, which was crucial for evaluating the long-term mechanical stability and durability of the films under operational conditions.

The remainder of the tests were carried out on non-aged samples. The testing involved assessing the hot-tack strength of the seals and determining tear resistance through the 'trouser' method. In addition, impact resistance was verified with a falling weight impact testing method, and the film-to-steel friction coefficient was determined. These studies were carried out only on films that hadn't undergone ageing, since the examined factors were crucial solely for packaging, and analysing aged films would not offer significant additional data. These test results validated that the film retains its mechanical integrity and is well-suited for use under challenging industrial conditions.

A key component of the experimental cycle in the doctoral research was microscopic examination, aimed at a detailed assessment of the morphology and structure of the film cross-sections. Examinations using optical microscopy revealed the film's three-layer configuration, with clearly defined boundaries between the individual layers. The SEM analysis of film fractures showed a consistent distribution of regranulate, calcium carbonate (with particle sizes not exceeding 24 µm), and titanium dioxide (a white pigment with particle dimensions from 200 to 600 nm) within the polymer matrix. It was demonstrated that the structure of layer B at the boundary with layer A predominantly exhibited crystalline fractures, whereas at the boundary with layer C, it often showed a localised fibrous structure with visible areas of plastic deformation around the filler particles. This indicates poor adhesion of the fillers to the polymer matrix, particularly in films containing 30% and 40% calcium carbonate. This effect is probably a result of the cooling method applied, which influences the structure of the films produced. The cooling of the formed film bubble was conducted from layer A towards layer C, significantly affecting the characteristics of the final structure.

SEM analysis also proved that incorporating regranulate with a higher flow rate into layer B did not cause the regranulate or calcium carbonate to migrate into the external layers. It was assumed that such migration could adversely affect the film’s functionality, but FTIR spectroscopy of the film's surface ruled out this possibility. Thermal properties and oxidative stability of the samples were evaluated using differential scanning calorimetry (DSC). These measurements allowed for the determination of the melting and crystallisation temperatures of the films under investigation. The DSC results indicated that the addition of regranulate slightly lowers the melting temperature of the films, without significantly affecting the functional properties of the material. DSC analysis also confirmed that the crystalline structure of the films remained largely unchanged despite the introduction of regranulate and calcium carbonate modifications. The crystallinity level of the modified films ranged from 33.63 to 40.82% (41.35% for the reference sample). In order to determine whether films containing regranulate and calcium carbonate are more prone to oxidation at high temperatures compared to the reference film and virgin film, oxidative resistance tests were conducted using the OOT method in an oxygen atmosphere. While the OOT tests demonstrated that the modified films have slightly lower oxidative resistance compared to the reference film, the difference was not significant enough to drastically affect the durability of the modified films in real-life applications, making them a suitable alternative to virgin films. Additionally, it's worth noting that the OOT results for the modified samples were more favourable compared to those obtained from the pure virgin LDPE granulate and the films made from it. "Even the lowest OOT temperature in the modified samples (EIV 604: 223.7°C) was higher than the OOT temperatures of the pure LDPE granulate and the films made from it, by 12.4°C and 10.6°C respectively. This further confirms that modifications with regranulate and calcium carbonate do not lead to significant thermal or oxidative degradation of the film.

In conclusion, the conducted research demonstrates that three-layer LDPE packaging films, produced with regranulate and calcium carbonate in the middle layer of the FFS three-layer film, retain their desirable mechanical and thermal properties while maintaining a consistent structure across the tested conditions. This makes them suitable substitutes for conventional FFS films made entirely from virgin plastics for industrial applications.