Recycling of flexible multilayer barrier films: structure–rheology–processing relationships and properties

Hissein Bechibo Adam[a], Mohamed Yousfi[a], Abderrahim Maazouz[a,b], Khalid Lamnawar*[a]

[a]*Université de Lyon, CNRS, UMR 5223, Ingénierie des Matériaux Polymères, Université Claude Bernard Lyon 1, INSA Lyon, Université Jean Monnet, F-69621 Villeurbanne Cedex, France*
[b]*Hassan II Academy of Science and Technology, Rabat, Morocco*

* khalid.lamnawar@insa-lyon.fr

Abstract :

For several decades, multilayer polymer films have attracted growing interest due to their versatility, lightness, and excellent barrier properties. These films, composed of two or more layers, exhibit a particularly advantageous synergy of properties. However, despite these assets, they remain difficult to recycle because of the incompatibility of the constituent materials. The present work aims to investigate the recycling of multilayer polymer films composed of polypropylene (PP), maleic anhydride grafted polypropylene (PP-g-MA) as a compatibilizer, and ethylene-vinyl alcohol (EVOH) as a barrier layer. The respective proportions of PP, PP-g-MA, and EVOH are 88%, 5%, and 7% by volume. This is a model study designed to explore the recycling of barrier multilayers of the PP/PP-g-MA/EVOH/PP-g-MA/PP type, under conditions where the composition is fully controlled, thus avoiding uncertainties and contamination associated with post-consumer waste streams. To this end, two recycling processes were investigated: mechanical recycling and selective dissolution–precipitation. In the first stage, the films obtained by co-extrusion were subjected to several cycles of mechanical recycling via twin-screw extrusion. Their recyclability was assessed by analyzing their thermal, rheological, and mechanical properties, as a function of processing parameters and the thermo-mechanical history they underwent. The results indicate that the thermal properties of the recycled blends are only slightly affected. In contrast, a significant decrease in viscoelastic properties, particularly viscosity, was observed up to the second cycle, followed by an increase at the third cycle, and then a further decline. This complex viscosity evolution can be attributed to several concomitant phenomena: PP β-scission, EVOH self-crosslinking, the continuation of copolymer formation at the PP-g-MA/EVOH interface, as well as phase separation induced by flow kinematics during mixing. Infrared (FTIR) and Raman spectroscopic analyses were systematically performed to probe and confirm these mechanisms. Furthermore, the addition of an additive (Nexamite R203) improved certain system properties. Model rheological studies in non-linear regimes, under shear and elongation, were also conducted. Elongational rheology proved particularly sensitive to the influence of these mechanisms on the macroscopic behavior, despite the low proportion of EVOH. The mechanical performance results corroborate these findings. In the second stage, the multilayer films were ground and then delaminated layer by layer using selective dissolution–precipitation. The isolated layers were characterized to evaluate their thermal and rheological properties. A slight degradation of these properties was observed, mainly attributable to solvent use during the separation process.

Keywords: multilayer films, recycling, extrusion, rheology, selective dissolution–precipitation

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