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Contribution of Thermal Modelisation to Understand adhesion Mechanisms in Extrusion Coating: Case of Polypropylene on Aluminium

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Abstract

A multilayered structure (polypropylene/grafted polypropylene/aluminium) manufactured by extrusion coating has been studied. The polypropylene film is extruded through a slit die, slightly stretched in air, coated on an aluminium foil in a laminator consisting of a chill roll and a flexible pressure roll, and finally cooled on successive chill rolls. Adhesive properties of polypropylene can be improved by grafting maleic anhydride on the polymer chain which may react with the aluminium surface. The reaction kinetic is more important for high temperature conditions.

The aim of this study is to characterize how process parameters can promote adhesion in multilayered structures made by extrusion coating. A model including crystallization has been developed to predict the temperature during stretching in the air and then during cooling on the chill roll. This model describes the film dimensions, the temperature along the process and especially where the polymer crystallizes. Results are in agreement with measurements of the surface temperature at different locations along the process line. Moreover, thermomechanical history has a real impact on structure and morphology in the film. All these aspects are revealed by microscopic observations of thin microtomed sections of the film and X-ray scattering. Finally, adhesion properties of laminate are linked with this thermomechanical history which depends on process parameters as polymer melt temperature, line speed and chill roll temperatures.

1 Thermomechanical model

After stretching in air, the polypropylene film geometry does not change any longer as soon as the molten film is coated onto the aluminium substrate. Consequently, the model can be divided into two parts: a thermomechanical model of stretching in air and a purely thermal model of cooling on the rolls.

The 1D-model used for stretching in air originates from previous works [1, 2] and takes into account cooling in air. It predicts the rate of stretching, the stretching distance as well as the width of the film and the temperature profile throughout the thickness. When polymer film is coated on the aluminium foil, the multilayered structure is cooled on several rolls and polymer layers crystallize. The energy balance equation for polymer has to take into account crystallization kinetic which is affected by the molecular orientation during stretching [3, 4].

Due to the succession of cooling rolls along the extrusion coating line, boundary conditions have to be adjusted carefully for each step: cooling in air, cooling in contact with rolls. Therefore, this work focuses particularly on the choice of appropriate boundary conditions, especially when a residual air gap is entrapped between the structure and the chill rolls because of a non-perfect contact.

2 Computation results

Stretching and cooling numerical models have been successfully compared to experimental temperature measurements in various processing conditions. As an example, Figs.1 show how important it is to take into account stretching effect on crystallization. If stretching effects are not accounted for (Fig.1 (a)), the model overestimates the film temperature which increases after cooling because polymer layers keep crystallizing in air. On the contrary, if they are introduced (Fig.1 (b)), the agreement is good.
The simulated thermal history allows to link the process parameters (temperature of the rolls, line speed) and the structure of the film revealed by microscopic observations and X-ray scattering.

**3 Correlation between adhesion and thermal history**

Our model can predict the thermal history at the polymer/aluminium interface up to the beginning of the crystallization. An important result is that adhesion is greatly affected by cooling conditions as the chemical reaction between grafted maleic anhydride and aluminium is promoted by high temperature conditions. Moreover, the reaction probability is mainly linked to molecular mobility at the interface which strongly decreases at low temperature or when the polymer crystallizes.

**4 Conclusions**

This study shows the interest to predict carefully the thermal history of a multilayered structure in order to understand adhesion mechanisms during extrusion coating. The general model of extrusion coating takes into account both stretching and cooling. It points out the impact of cooling conditions on the crystalline structure of the polymer layer and on adhesive strengths. It appears that adhesion properties are very sensitive to high temperatures or low cooling rates which are favourable to the reaction between grafted polypropylene chains and aluminium foil. Consequently, cooling process and polymer properties (molecular mobility, relaxation time and crystallization rate) are the key points of extrusion coating for an optimization of adhesion properties of polypropylene/aluminium structure.

**5 References**