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Chemical Modification of Polyolefins for Improved Adhesion by Reactive Extrusion

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Grafted polymers are of great interest for industrial applications and fundamental research mostly due to their improved properties and specific characteristics compared to the pristine polymer.[1] Applications for grafted polymers have emerged in various sectors, such as packaging, drug delivery, hydrogels, stimuli-responsive polymer materials, surface modifications, and coatings.[2] A common application is the use as compatibility layer between two different materials in foil extrusion, or directly as adhesion layer.[3] Here, maleic anhydride is typically used as monomer to graft to polyethylene or polypropylene.[4] Molecular interactions are enabled due to the polar and reactive anhydride group, which provide enhanced adhesion to different substrates.

However, many grafting processes lack in efficiency and therefore in industrial applicability. Reactive extrusion is a straightforward one-step grafting technology, in which the grafting reaction proceeds in the extruder barrel and the desired polymer is directly obtained. The challenge in reactive extrusion technology is that the reaction has to be completed within the very short processing time. Thus, kinetics need to be analysed and fitted to the extrusion process.[5]

A twin screw extruder with screw diameter of 25 mm was used for the grafting reaction of triethoxyvinylsilane to very low density polyethylene (VLDPE). The extruder consisted of five heating zones; thereof the three middle ones were equipped with an air cooler. The feeder section was water-cooled. The screw design is shown in Fig. 1.

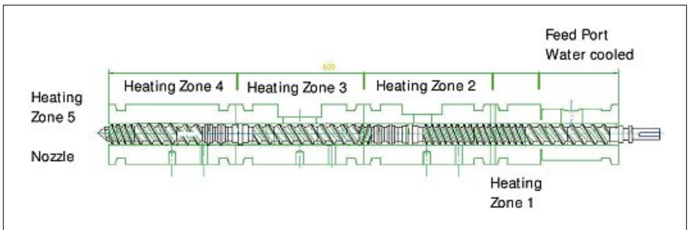

Fig. 1. Screw design of the used extruder having five heating sections.

Triethoxyvinylsilane was used as grafting agent since silanes are important adhesion promoters. The residential time of the polymer in the extruder was determined by adding graphite and measuring the absorption with a photospectrometer revealing a residence time of around 150 s after a delay of 100 s (see Fig. 2).

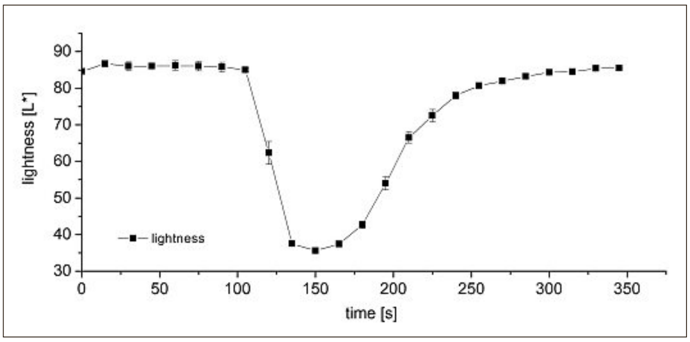

Fig. 2. Residence time graph of VLDPE at 180 °C with a mass flow of 2 kg/h and a screw speed of 80 rpm.

The process was thoroughly investigated by IR spectroscopy, headspace GC-MS and rheology measurements. The combination of these three analytical techniques proved to be a powerful analysis tool box for the reactive extrusion process.

Different initiators were screened and evaluated for their best performance. Grafting efficiency in dependence of temperature, initiator type, initiator concentration, and thermal stabilizer were analysed. After characterization and optimization of the reactive extrusion process, T-peel tests on thin aluminum substrates were performed to verify the adhesion behavior. To our delight, the modified VLDPE adhesives clearly showed improved adhesion on aluminum. While non-modified VLDPE exhibits mostly no adhesion on the substrate, the T-Peel strength of modified VLDPE was in the range of 0.6–2.6 N/mm.[6]

Overall, triethoxyvinylsilane was successfully grafted to VLDPE by reactive extrusion and greatly improved adhesion to aluminium could be obtained.

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