

## 5 CONCLUSIONS

The extrusion trials done using the Berstorff twin-screw extruder and the Rapra single-screw extruder illustrate that the conversion of granular starch into thermoplastic starch requires gradual application of shear in the presence of high plasticiser contents. Excessive shear is harmful to the starch and results in embrittlement due to a reduction in the average molar mass. Every extruder is unique and therefore processing parameters, barrel temperatures, screw speeds, etc. should be set up by systematic experimentation.

Initial extrusion and moulding trials revealed that the TPS compounds were very difficult to process. Difficulties were encountered with feeding the dry blends into the compounding extruder. The addition of 2,5% precipitated silica was necessary to facilitate feeding of the dry blends into compounding extruder.

The compounding processing window is relatively small. Only formulations within a narrow range of water and glycerol content were extrudable. All TPS compounds showed poor processability during injection moulding, especially in the sprue part. This was improved by adding stearyl alcohol at ca. 1,5% as an external lubricant and mould-release agent. Nevertheless, for some compositions it was also necessary to use “Spray-and-Cook” as mould-release agent during injection moulding.

Good mechanical properties were only achieved by using high-amylose starch. Starch high in amylopectin was very difficult to process and showed excessive retrogradation, with samples even cracking spontaneously during ageing. Increasing the glycerol content increased the elongation-to-break but reduced the tensile strength of TPS compounds.

The TPS-PVB blends showed highly non-linear composition-dependence. Scanning electron microscopy (SEM) and dynamic mechanical analysis (DMA) revealed a phase-separated nature for all the TPS-PVB blend compositions investigated. The tensile properties were negatively affected by ageing in a high-humidity environment and they deteriorated rapidly when the samples were soaked in water. Synergistic property enhancement was observed for a compound containing 22% thermoplastic starch. It featured a higher tensile strength, showed better water resistance and was significantly less affected by ageing. At higher PVB levels, the property dropped to values that were lower than expected from the linear blending rule.



Blending the TPS with polyamides improved the processability and also the mechanical properties. Blends with polyamide EMS were found to have reasonably good mechanical properties. However, starch at low levels did not provide the polyamides with a reinforcing effect. The phase separation during extrusion was caused by the difference in glass transition temperatures, resulting in poor tensile properties for blends containing more than 50% Euremelt. The properties of all the compounds investigated were affected by moisture content and also by ageing.

Blends with recycled plasticised PVB showed highly non-linear mechanical property variations with composition. The best was a compound containing approximately 22% TPS. However, DMA, enzyme erosion and SEM experiments showed that all the TPS-PVB blends had a phase-separated morphology. It is concluded that, at low to medium starch content, the starch acts as reinforcing filler in the PVB matrix. The deterioration in mechanical properties at higher starch levels is attributed to the starch becoming the continuous phase.

The two-phase nature of the TPS-PVB blends is not surprising as most polymer pairs are thermodynamically immiscible [Paul and Newman, 1978]. This phase separation leads to the creation of internal interfaces. If these have a high interfacial tension, there will be poor adhesion between the two phases. The end-result is poor stress transfer between phases and a loss in mechanical properties, as observed in the present case for the blends with high TPS content [Paul and Newman, 1978].

The blends rich in PVB show better mechanical properties, especially the compound with 22% TPS. Although this blend is also phase-separated, there appears to be sufficient interaction between the polymer pairs at the molecular level to induce mechanical “compatibility”, rather than miscibility. For this blend the stiffer starch domains appear to provide some reinforcing effect, leading to an increase in the tensile strength and modulus. Furthermore, the SEM evidence suggests that debonding at the interfaces occurs only after some plastic deformation of the matrix has occurred. This internal energy-absorption process explains the increase in the measured elongation-to-break for this sample.