# STARCH – POLYVINYLALCOHOL – ACETAL : A WATERSOLUBLE FILM

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Abstract: To ensure more comfort, dosage units of active ingredients may be packed in water soluble film prior to application. In the mean time plasticised polyvinylalcohol (PVAL), with limited metabolitic degradability in common water epuration units, is used for present purpose. In this work we found that the reducing chain ends of degraded starch molecules form semi acetals with the hydroxyl moieties of (PVAL). Reaction conditions could be selected for processing in a twin screw extruder. Due to replacement of around 50% of the polyvinylalcohol by starch the production costs are significantly lower than that of pure PVAL-films. The acetal films show better mechanical properties, seal easier, dissolve faster in water and mineralise at lower temperatures if compared with pure PVAL.

## CHEMISTRY:

To raise the concentration of the reducing end groups of the poly 1,4 (1,6) anhydroglucose chains, native starch was treated with aqueous mineral acids (Ref. 1). The chain scission process was controlled by the titration of the reducing end groups with a modified Fehling reagent (Ref. 2). Products with a weight average degree of polymerisation around 10 were selected for the acetalisation reaction.

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The course of degradation was followed by the measurement of the limiting viscosity:

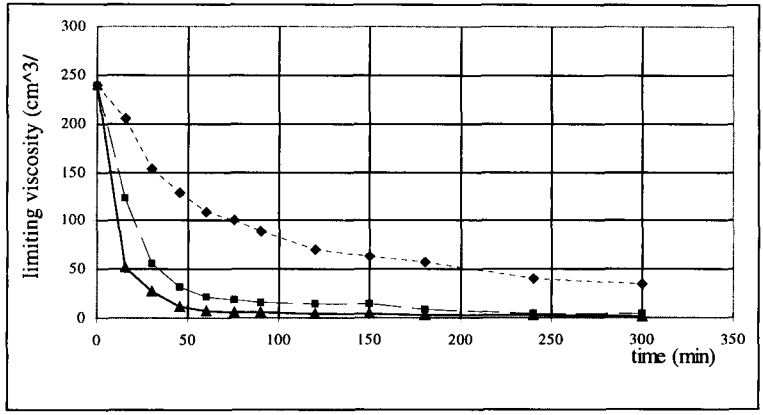


Figure 1: the limiting viscosity plotted against duration of degradation of starch with aqueous HCL : ▲0,1 N ; ■0,05 N ; ◆0,01 N at 120°C.

The acetalisation of PVAL with dextrin was acid catalysied using sulphuric acid in dimethylsulfoxide as solvent (Ref. 3). The reaction mixture was dialysed against water, dried and dissolved in dimethylformamide. The solvent was distilled off and the dried product was analysed.

The content of anhydroglucose in the reaction product was detected by solid state 13C-NMR spectroscopy:

The relative intensity of the signals at 20 and 102 ppm chemical shift allowed the determination of the anhydroglucose content.

The turn over of the acetalisation reaction was determined at different reaction times and temperatures:

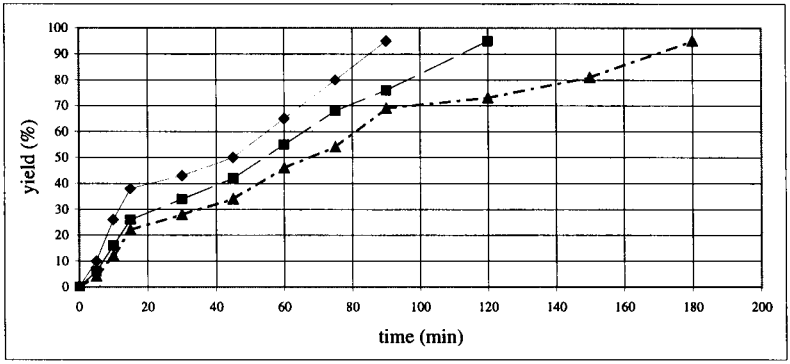


Figure 2: Kinetics of the acetalisation of PVAL with dextrin in dilute DMSO solution at: ▲ 80 °C; ■ 110 °C; ● 140 °C.

To reduce the necessary reaction time the reaction was repeated in molten starch / PVAL plasticised with 20 % water.

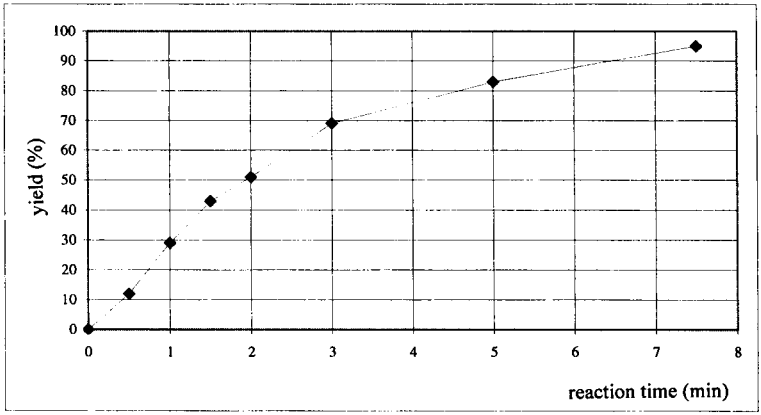


Figure 3: Kinetics of the acetalisation of PVAL with dextrin in the melt plasticised with 20 % water at 110 °C.

The reaction time could be reduced in the melt by more than a decade due to the fact that the reaction is diffusion controlled. Thourough mixing of the melt during the reaction was facilitated in a corotating twin screw extruder.

The reaction product was granulated and processed in a film blowing unit with the following parameters:

melt-temperature : 135 °C; melt extension ratio in the direction of extrusion 1:4; perpendicular to the direction of extrusion 1:4; final film thickness 25 µm.

MECHANICAL PROPERTIES OF STARCH-PVAL-COPOLYMER FILM

*simple stress - strain behaviour of the blown film:*

| material | thickness in µm | Youngs Modulus (MPa) | Stress at Peak (MPa) | % strain at yield (MPa) | solution time in water (sec) |
|---|---|---|---|---|---|
| | | | | | |
| pval 1 | 40 | 1894 | 32 | 72 | 15 |
| pval 2 | 40 | 2025 | 36 | 62 | 30 |
| pval 3 | 40 | 1936 | 32 | 70 | 90 |
| pval-dextrin-copolymer | 45 | 326 | 26 | 80 | 15 |

Stress-strain behaviour and the solution time was measured at 20 °C and the speed of elongation was 10 cm / min.

Biological degradability detected with the OECD 301 test.

| material | Degradation after 28 d at 12 °C (%) | Degradation after 28 d at 25 °C (%) |
|---|---|---|
| | | |
| pval 1 | 0-3 | >80 |
| pval-dextrin-coploymer | >60 | 100 |

## PRESENT STATE

Production trials and market introduction. Patents are pending.

## ACKNOWLEDGEMENT

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