

NATURAL FILLERS REINFORCED THERMOPLASTIC MAIZE STARCH

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There is an increasing interest in nonfood uses of starch such as the substitution of synthetic polymers because of its total biodegradability and worldwide availability at low cost. Native starch can be transformed into a thermoplastic material (TPS) through thermo-mechanical treatment in the presence of suitable plasticizers, such as water and glycerol. However TPS has two main drawbacks: it is highly hydrophilic and mostly water-soluble and it has poor mechanical properties, the reinforcement with organic filler is an available options to overcome these weaknesses.

The aim of this research is to prepare thermoplastic starch based composites with organic natural fillers in order to achieve good mechanical properties maintaining, however, biodegradability. The effect of two different plasticizer (glycerol and sorbitol) and of two natural fillers (Flax, Cellulose) was investigated and compared with a classical mineral filler (Talc).

The starch used was the maize starch CERESTAR RG 03408 (Cargill). Glycerol ReagentPlus with purity $\geq 99,0\%$ (Aldrich) and Sorbitol (Aldrich) were used as plasticizer for starch. Cellulose, flax fibers and talc (HTP1 from Imifabi) were used as fillers.

The samples for Dynamic-mechanical thermal analysis (DMTA) were prepared by hot compression molding (5 MPa) at 160 °C for 3 min. The temperature range was from 30 °C to 120 °C, heating rate 3 °C/min. The tests were performed after 15 days conditioning at 25 °C and 50% RH.

In a first step TPS was obtained by a twin co-rotating extruder ($\Phi=18$ mm). The starch was dried for 5 hours at 120 °C in a vacuum (-1000 mbar) oven thus it was mixed manually with plasticizers. During the extrusion process a temperature profile from 115 to 150 °C was used with a screw speed of 150 rpm and a flow of 1kg/h. The ratio of plasticizers for glycerol and for sorbitol were respectively 30% and 40% wt/wt. In a second step all the materials were mixed and fed through the main hopper of a co-rotating twin screw micro extruder. Residence time was fixed for all runs at 10 minutes. The screw speed was fixed at 100 rpm for the melt mixing. The heating temperature 160 °C.

The nature of the plasticizer influence the final performance of the material. The mechanical properties of starch plasticized with glycerol (TPSG) are very low and sensible to moisture thus it is unsuitable for the production of plastics. The addition of sorbitol to starch (TPSS) allows a good plasticization and mechanical properties are good also after conditioning: the storage modulus is near to common polyolefins. The sorbitol is thus the better material for plasticizing starch if mechanical properties are needed. Unfortunately TPSS mechanical properties strongly decreases with temperature.

Natural organic fillers improve mechanical properties of the two TPS. The addition of 20 wt% cellulose in TPSG matrix results in a storage modulus increase of more than 300%. The same behavior is achieved adding 30 wt% of flax on the contrary only a little improvement is obtained with 30 wt% of talc.

In the case of TPSS adding cellulose in the same content shows a stronger storage modulus increase at 35 °C (about 400%). A considerable improvement of rigidity was instead observed at higher temperatures (about 14 times). A similar behaviour was observed with flax even if the reinforcement effect is less high (300% at 35 °C and 1000% at 85 °C). Talc is not so effective as other used fillers ("only" 300% of increase). Probably the stress transfers at the filler/matrix interface is better with TPSS and organic filler thus this gives a better mechanical properties even at high temperatures.

Table 1: Storage modulus of thermoplastic starch composites based on TPSG and TPSS.

Matrix Filler [wt %]	TPSG				TPSS			
	0%	Cell20%	Flax30%	Talc30%	0%	Cell20%	Flax30%	Talc30%
Modulus @35°C	60	200	200	118	460	2000	1500	1270
Modulus @85°C	23	60	64	22	25	350	270	88