# LOW TEMPERATURE STORAGE OF THERMOPLASTIC STARCH

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**Summary.** The paper presents the results of studies on the mechanical properties of thermoplastic starch (TPS) mouldings stored under low temperature. An influence was examined of freezing and chilling conditions on the selected properties of samples measured on the strength appliance Zwick type BDO-FBO 0.5 TH

Key words: thermoplastic starch, extrusion, mechanical properties, injection moulding machine

### INTRODUCTION

Thermoplastic starch (TPS) can be produced in machines and equipment designed for synthetic polymers processing. The biopolymers may be an alternative supplement to the existing synthetic products. However, starch plastics are not widely introduced as consumer goods due to some major drawbacks. Many publications have appeared concerning thermoplastic starch application in the injection moulding process. In the eighties the Capsugel Society patented a method for the production of starchy capsule for drugs. In 1984 the first patent was presented describing the process of injection moulding [Onteniente et al. 2000]. Special emphasis was put on the injection pressure. This patent, though, does not state explicitly what injection parameters should be employed and if the process is fully automatic. The major drawback in the injection moulding is the primary shrinkage of a product. To improve the biopolymer mechanical properties, it should be comparable to some natural fibres, like flax, hamp and jute in the amount up to even 20-45% [Smits 1999, Sinha Ray 2005, Wollerdorfer, Bader, 1998]. The fibre additive improves the structural properties of a material even by a few times. The packages produced from this type of material should be characterized by strength properties of the traditional plastic, yet after having been used it should be compostable [Fritz et al. 2001, Funke 1998, Leszczyński 1999].

In our researches special concern was given to the issues of production and physical properties of test bars obtained by means of injection moulding technique with the application of TPS granulates with differing content of a plasticizer and natural fibres, stored under low temperature. As a potential packaging material TPS had to be used in different storage conditions.

## MATERIALS AND METHODS

The raw material used for the tests was commercial potato starch produced by Avebe b.v. (NL) blended with glycerol at varied proportions. Glycerol (98.5% purity) came from the Netherlands. There were also used cut flax and cellulose fibres supplied by a Polish producer.

The total process consisted of two steps. In the first step starch was gelatinized, mixed with glycerine in a twin screw extruder and pelleted. For the experiments with fibre reinforcement the flax or cellulose fibres were mixed into the starch during the extrusion. The pellets are very similar to pellets in plastic processing and they can be readily fed to the second step where the product obtains its final shape. This second step was injection moulding (to form 3-dimensional objects).

The samples of 10 kg ware prepared by means of the laboratory strip mixture from the mix of starch of 16% humidity and glycerine. The effective mixing time was set to 20 minutes. The glycerine share was from 22% of the mixture weight, the fibre share was, for selected mixtures, from 5% to 10%. After mixing, the samples were left in airtight plastic bags for 24 hours in order to cause an intensification of glycerine penetration into the starch granules. Directly before the extrusion, the mixtures were stirred again for 10 minutes in order to guarantee the effect of loose and slack mixture structure and assure a regular feed in the subsequent mixing process [Janssen, Mościcki 2006, Oniszczuk 2006].

Extrusion of biopolymer was performed on a co-rotating twin-screw extruder by PASQUET-TI, an Italian production, characterized by L/D=5, and the screws diameter -45 mm.

The granulate was made at one speed of the extruders screws, that was 70 rpm. The extruder's head was fitted with a bronze matrix having one hole of the diameter  $\phi = 3$ mm. The product was treated with a high-speed knife, which helped to obtain granulate of the previously-set small size. The temperature of extrusion in particular extruder's parts and head was between 80°C and 140°C; pressure in the head fluctuated between 10 MPa and 18 MPa. The obtained granulate was dried in a vacuous laboratory drier at the temperature of 50°C for 6 hours until the humidity of 4-6% was achieved (Fig. 1.).



Fig. 1. Granulates with a) 10%, b) 5% content of flax fibres and c) 5% content of cellulose fibres

Different batches of granulate were produced: without and with addition of fibres in different proportions. The final moisture content of granulates was measured after the product had been formed. Final TPS processing was performed on the injection moulding machine ARBURG 220H90-350, L/D = 20.5. The injection speed was maintained at the level of 70-90 mm/s, injection time 5s, and the temperature of the processes was 140°C.

To obtain the samples an injection mould was used which produced the basic profile in the form of a 'spade' used during further examination of mechanical properties of mouldings (Fig. 2.).



Fig. 2. Injection mould of the injection machine ARBURG 220H90-350

The examination of the mechanical properties of biopolymer mouldings was performed on a strength appliance machine-Zwick type BDO-FBO 0.5 TH equipped in the head 0.5 kN, moved with the speed of 3 mm/min. The test focused on the maximum stress and maximum elongation during stretching.

In order to examine the behaviour of products in low temperatures, the mouldings were placed in a refrigerator by POLAR type CP 1116A (chilling), and a freezer by Whirlpool type AFG 6450 AP. Samples were put in single layers and stored at two different temperatures: +3 °C and -36 °C for 24 hours at a given stage and for 30 days. After taking out, the samples were placed in heatinsulating containers (in order to avoid moulding temperature fluctuation), and subsequently were individually put to strength tests. These tests gave an indication of the quality of the packaging at material in cold – storage conditions [Oniszczuk 2006, Van Soest 1996].

#### RESULTS

Starch biopolymers may be utilized in packaging industry not only as protective wrappings for products stored in ambient conditions, but also in low temperatures and frozen. In connection with the possible applications of starch biopolymers, for example, as ice-cream sticks or stiff packaging for frozen food, selected mechanical features of starch mouldings were tested after storage in varied temperature conditions. To illustrate the performed testing, the mouldings produced at 140°C were used. The measurement results are presented in Figure 3. Mouldings produced in such conditions had varied mechanical strength depending on the used additives and storage conditions.

Mouldings produced without the addition of fibres displayed low stress values in the temperature lower that ambient temperature.

Mouldings with varied addition of flax fibres had similar mechanical strength in ambient temperature. 24-hour storage in low temperatures considerably changes the sample strength. Surprisingly, the mechanical strength level of mouldings with flax fibres was higher than that of the other tested mouldings, which might be an effect of low temperature on the interaction between starch, glycerine and flax fibres, and this, in turn, caused an improvement of moulding strength. Higher addition of flax fibres (10%) contributed to the most improved strength among all the tested mouldings during the break test. (Fig.3). For mouldings with cellulose fibres, higher stress values were also recorded after cool storage, as compared to mouldings without additions. However, the values of sample stress did not exceed 20 MPa, and the highest was set at the storage temperature of  $- 36^{\circ}$ C



Fig. 3. Influence of storage temperature and the content of natural fibres in mouldings on its behaviour during tension test (glycerine content 22%, storage time 24h)

The preparation time extended to 4 weeks did not cause significant changes in the strength of the tested products obtained at 140°C without and with the addition of plant fibres. In mouldings with no fibre addition lowered mechanical strength was observed at the temperature of + 3°C, and storage at - 36°C had no impact on sample strength. During this examination the highest mechanical strength was recorded for mouldings with an addition of flax fibres, regardless of temperature of storage (Fig. 4). In freezing conditions mouldings with these fibres were the most stable with prolonged storage and sample strength remaining unchanged (about 27 MPa). Almost identical values were observed for mouldings with cellulose fibres which, after long storage both in cool temperature and frozen, exhibited high stability of the tested parameters.

The results of measurement of plasticity as the parameter of resistance to deformation during storage for 24 h are shown in Fig. 5. The top plasticity regardless of temperature of storage was recorded for mouldings produced without an addition of fibres in the full range of applied temperatures of production. When testing mouldings with an addition of flax fibres it was observed that the material stored in cool conditions displayed better plasticity. Elongation values were reduced for

samples containing 10% of flax fibres. A reverse relationship was observed for mouldings having cellulose fibres. The lowest elongation and, consequently, the best utilitarian features were displayed by mouldings with 5% of cellulose fibres stored in  $+ 3^{\circ}$ C, both after 24 h and after 4 weeks (Fig. 6). Generally, it can be concluded that the lengthening of storage time to 4 weeks did not particularly influence the elasticity of mouldings both with fibres and without.



Fig. 4. Influence of storage conditions and fibres content in mouldings on its behaviour during break test (glycerine content 22%, storage time 4 weeks)



Fig. 5. Influence of storage temperature and the content of fibres on its behavior during tension test (glycerine content 22%, storage time 24 h)



Fig. 6. Influence of storage temperature and content of fibres in mouldings on the size of maximum elongation of samples during tension test (glycerine content 22%, storage time 4 weeks)

#### CONCLUSIONS

The storage of biopolymers in low temperatures differently and significantly influenced their mechanical properties (maximum stress, maximum elongation).

Biopolymers produced from starch might be successfully used for the manufacturing of packaging for food subjected to cool storage. Temperatures (+3 °C, -36°C) and sample storage time (24 h and 4 weeks) had no significant influence on the quality parameters of biopolymers.

In the full range of cool storage temperatures, the best usage parameters were displayed by mouldings containing 10% of flax fibres. This biopolymer should find broad application in the packaging industry and, in particular, in the manufacturing of disposable trays used in food subjected to low temperature storage.

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## WPŁYW NISKICH TEMPERATUR NA PRZECHOWYWANIE SKROBI TERMO-PLASTYCZNEJ

**Streszczenie.** W pracy przedstawiono wyniki badań właściwości mechanicznych wyprasek wyprodukowanych ze skrobi termoplastycznej (TPS) przechowywanych w niskich temperaturach. Badany był wpływ warunków zamrażalniczych i chłodniczych na wybrane właściwości mechaniczne próbek. Testy wytrzymałościowe przeprowadzono przy zastosowaniu maszyny wytrzymałościowej Zwick typ BDO-FBO 0,5TH.

Słowa kluczowe: skrobia termoplastyczna, ekstruzja, właściwości mechaniczne, wtryskarka