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Extrusion-cooking of starch protective loose-fill foams

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ABSTRACT

One of the most interesting alternatives to EPS is extrusion of starch-based materials. TPS-based biocomposites can be processed, in a one-step process, via an extrusion-cooking. Wide program of the experimental works with application of extrusion-cooking for production of starchy loose-fill foams has been started in the Department of Food Process Eng., Lublin University of Life Sciences in 2012. The object of the study is to achieve commercially acceptable biodegradable products based on locally produced potato, corn and wheat starch, which can replace popular EPS. Results of the first phase of the study are presented in the paper. The measurements of glass transition temperature of TPS samples showed that with glycerol content growth in the blend, the *Tg* of the obtained material decreases almost linearly. In the case of potato TPS, the highest observed *Tg* was 187.7 °C for 7.0% glycerol and the lowest was at 18.1 °C for 30% glycerol. Properties of the loose-fill foams highly depend on raw materials and process parameters used in production. In addition of plasticiser or other additives, different temperature of processing is causing changes in product's properties. All starch-based foams had high open-cell content and the expansion was attributable to the escape of water as steam during processing, resulting more than 80% open cells. The foam density of starch-based products ranged between 18.7 and 30.5 kg/m^3. The products were at least 2.5 times more dense than EPS-based foams. The best products achieved by us up till now are the corn TPS-based foams containing 3% plasticizer and 1% poly-vinyl alcohol. The energy consumption during extrusion-cooking depended on the material composition of the blends, temperature and the screw rotation speed used during processing. Average value of SME was about 2.52×10^5 J/kg, which is equivalent to 0.07 kW h/kg.

Keywords: Extrusion-cooking; Thermoplastic starch; Starch foams; Glass transition; Physical properties

1. Introduction

Expanded polystyrene loose-fill foam products (EPS) are commonly used to provide cushioning, protection and stabilization of articles packaged for shipping. However, their application has enjoyed a steady growth over the last decades, creating a great environmental problem due to poor compostability being practically non-biodegradable. Million tons of plastic retail bags, packaging film, shaped solid boxes and loose fillers are thrown into garbage daily, resulting in a lively debate as to how to replace them to reduce their negative impact on the environment. One of the most interesting alternatives to EPS is extrusion of starch-based materials. Several patents on the extruded foams based on starch and blends of starch with various functional additives have been published (Bastioli et al., 1998a,b; Belloti et al., 2000; Xu and Doane, 1998) and nowadays such materials are commercially available. The influence of extrusion conditions, moisture content and composition of the blends used on the physical properties of starch-based foams have been reported (Bhatnagar and Hanna, 1995; Tatarka and Cunningham, 1998). Many R&D centres, especially, those that are packaging producers are busy in recent years introducing new products in the market. Various synthetic polymers like poly(vinyl alcohol) or

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polycaprolactone, have been blended with unmodified starches to produce foams with lower densities and increased water resistance.

1.1. *Thermoplastic starch (TPS)*

The target of recent investigations is to obtain commercial packaging material produced from pure starch and to exclude synthetic polymers from the formulation. TPS seems to be a perfect solution because it can be processed with conventional technologies used in synthetic plastic manufacture (extrusion, injection moulding) (Shogren et al., 1993; Wiedmann and Strobel, 1991; Janssen and Moscicki, 2009). To obtain TPS, thermal and mechanical processing should disrupt semi crystalline starch granules. As the melting temperature of pure starch is substantially higher than its decomposition temperature, there is a necessity to use plasticizers like glycerol. Influence of both, temperature and shear forces, create the disruption of the natural crystalline structure of starch granules; and polysaccharides form a continuous polymer phase (Avérous et al., 2001; Nashed et al., 2003; Shogren et al., 1993; Van Soest et al., 1996). If the total thermal and mechanical energy provided to the starch is insufficient, the product will show unmolten starch granules of clear crystallographic structure. Similarly, an insufficient amount of plasticizer may result in an incomplete destruction of the crystallographic structure of starch (Souza and Andrade, 2002; Van Soest and Knooren, 1997). To improve the mechanical properties of TPS based materials, many different additives can be applied, like emulsifiers, cellulose, plant fibres, bark, kaolin, pectin and others (Avérous et al., 2001; Ge et al., 2000).

It is well known that starchy materials are brittle. This is related to a relatively high glass transition temperature T_g - the most important parameter for determining the mechanical properties of amorphous polymers and for controlling of their crystallisation process (De Graaf et al., 2003; Myllärinen et al., 2002; Zeleznak and Hoseney, 1987). The method of differential scanning calorimetry (DSC) is used most commonly to determine the glass transition temperature while other methods are NMR (nuclear magnetic resonance) or DMTA (dynamic mechanical thermal analysis). The great importance lies in the starch composition (amylose and amylopectin ratio) during processing. Influence of water on the T_g showed that the very branched amylopectin had a slightly lower glass transition temperature than the amylase and starchy material containing water that is generally in the glassy state, and therefore brittle under natural conditions (De Graaf et al., 2003).

The results of measurements published by various authors differ substantially due to different measurement process conditions used and complex changes that occur in starch during thermal treatment. Myllarinen et al. (2002) indicated that T_g of amylose and amylopectin may equal the room temperature when the water content in a blend is 21%; however, at the same glycerol content, it goes up to 93 °C. Yu et al. (1998) hold that TP maize starch with 10% moisture and 25–35% glycerol shows a T_g running from 83 to–71 °C. Van Soest and Knooren (1997) proved that potato TPS with 11% moisture and 26% glycerol had a $T_g = 40$ °C; whereas, for the materials of higher moisture and glycerol content, it fell below 20 °C. Lourdin et al. (1997) reported that potato starch of 13% moisture with 15% glycerol content had the T_g around 25 °C, while at 25% glycerol, the T_g dropped to around 0 °C.

The TPS mechanical properties depend on the temperature of starch processing, water content as well as quantity and type of added plasticizers and aid materials. Increase of the plasticizer content brings about a decrease in tensile strength of TPS; whereas, the elongation at break increases. Starch is a natural polymer containing numerous hydrogen bonds between the hydroxyl radicals in its molecules; therefore it manifests substantial tensile strength values. Glycerol, sorbitol or glycol behave like diluents and decrease the interaction between molecules and consequently, they diminish tensile strength. At the same time, they act as placticizers that improve macromolecular mobility and leads to a rise in elongation at break (Liu et al., 2001; Shogren, 1993; Yu et al., 1998). Addition of filler materials like cellulose fibres, flax, kaolin or pectin increases the tensile strength but decreases the elongation at break. In turn, urea or boric acid addition improves the elongation at break but decreases the tensile strength (Fishman et al., 2000; You et al., 2003; Yu et al., 1998).

During storage of TPS, some recrystallisation of amylose and amylopectin occurs. Together with a longer storage period and consequently with a crystallinity of TPS; tensile strength increases and elongation at break decreases. The increase of moisture content of the starchy materials storage conditions brings about intensification of their mechanical properties changes (Van Soest and Knooren, 1997).

1.2. *Processing of TPS*

TPS based biocomposites can be processed, in a one-step process, via an extrusion-cooking (Janssen and Moscicki, 2009). Generally speaking, extrusion-cooking of vegetable raw materials (technique widely used in food sector) consists in the extrusion of grinded material at baro-thermal conditions. With the help of shear energy exerted by the rotating screw and additional heating by the barrel, the food material is heated to its melting point or plasticating-point (Moscicki and Van Zuilichem, 2011). In this changed rheological status, the food is conveyed under high pressure through a die or a series of dies, and the product expands to its final shape. This results in more different physical and chemical properties of the extrudates in comparison to raw materials used.

Food extruders perform cooking tasks under high pressure. Physical– technological aspects like heat transfer, mass transfer, momentum transfer, residence time and residence–time distribution have a strong impact on the food properties during extrusion-cooking, and can drastically influence the final product quality. An extrusion-cooker is a process reactor in which the designer has created the prerequisites in the presence of a certain screw lay-out, the use of mixing elements, the clearances in the gaps, the installed motor power and the barrel heating and cooking capacity, to control a food and feed reaction. This can be a reason "in itself", when only mass is transferred in wanted and unwanted reaction products due to heating, e.g. the denaturation of proteins under presence of water and the rupture of starches are both affected by the combined effects of heat and shear. The reaction can also be provoked by the presence of a distinct biochemical or chemical component like an enzyme or a pH controlling agent (Moscicki and Van Zuilichem, 2011).

Application of extrusion-cooking technique to process starch-plasticizer mixtures can be one of the most economical and efficient way to produce TPS loose-fill foams. The process conditions are to be very stable and strictly determined according to the expected quality of the extrudates. Also, raw materials used (starchy components mixed with the plasticizers) have to be fitted properly. They all need many