

Application of extrusion-cooking technique for foamed starch-based materials

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Abstract. Foamed materials are widely used, mainly as a protection objects during transport of various products. Traditionally foams are produced from plastics so they are very difficult for waste management. It is the challenge for many scientific centres to develop a technology for the production of bio-based materials which can be rapidly decomposed. The task for the researcher is to obtain a relatively cheap, easy to use and completely biodegradable materials. The aim of this work was the selection of the main raw materials, functional additives and process parameters to obtain the most effective parameters of extrusion-cooking process for foamed starch-based materials. Properties of the products and processing costs were taken into account. During the study, the extrusion-cooking process was performed under various conditions: temperature, humidity, type of the die, screw rotational speed, various raw materials and additives blends. The best results were obtained for mixtures based on potato starch and with addition the foaming agent Plastron foam PDE and poly(vinyl) alcohol PVA.

1 Introduction

Producers of foamed packaging materials are under increasing pressure of law regulations concerning environmental protection and waste disposal [1-4]. Packaging materials are a serious problem for businesses and local community, because their waste are difficult to manage and decompose in natural environment. Also recycling process is often unprofitable (too expensive transport and handling costs). More and more attention is being paid to the possibility of reprocessing of packaging materials, especially one-time use. The market is starting to require from manufacturers environmentally safe materials which have obviously the utility requirements. The interest in environmentally friendly materials stimulates the development of the production of raw materials from natural raw materials for packaging purposes[5,6].

Currently it is not possible to reuse all produced materials based on plastics. Hence, attempts were made to use agricultural crops in the production of biopolymers [7-10]. It is possible to use starch, protein or cellulose in the processing of biodegradable packaging materials. Biopolymers based on starch and cellulose have the great advantage – they are suitable for composting or reuse. A successful substitution of polymer-based porous materials with starch-based materials is being sought.

Starch-based foamed materials are lightweight and can be used successfully in packaging [11-14]. However biodegradable foamed materials, based on natural raw

materials, are characterized by brittleness and poor water resistance. These materials dissolve completely, as opposed to synthetic foams which are neutral to moisture [15-19]. The additional disadvantage of this type of materials is the difficulty and limitations in creating the optimal cell-like structures which are required by potential customers. The lack of industry-ready solutions is a challenge for research institutions to obtain the possibility of producing fully functional, biodegradable foamed materials that can be widely used in packaging production.

Extrusion-cooking is a process of extrusion of loose material (mostly raw origin materials) under the influence of high pressure and high temperature [20-25]. It leads to the development of significant physical and chemical changes in the processed material. The process is carried out in devices called extruder-cookers [26-28]. The main working part of each machine is a screw or a pair of screws (hence the division into single and twin screw extruders), which are located in a cylinder (special casing). During the extrusion-cooking process working part squeezes the raw material and extruded it through a die installed at the end of the cylinder. Extrusion-cooking technology is widely used in agri-food processing because of its versatility and very high efficiency.

The extrusion-cooking process can be used to mold plastic with the addition of natural starch. As it was said in many research, starch can be a component of biodegradable materials and be exist in native or extruded form [29-34]. Starch which is processed by

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extrusion-cooking goes into destructuring and has gelatinise form. It has thermoplastic properties which allows starch to be molded and blended with other components [35-38], including traditional polymers. In relation to native starch, the extruded version has other rheological properties and is more sensitive to water. The properties of starch can be created by selecting the appropriate extrusion-cooking process parameters, as well as the selection of natural raw materials with the required properties and the use of various functional substances and functional ingredients. Synthetic materials with added starch are primarily used for the production of films, containers and foams. The thermoplastic starch obtained by extrusion-cooking is referred to as TPS. The advantage of using extruded-cooking starch in packaging is its cost and price of ready to use material.

The aim of this work was the selection of the main raw materials, functional additives and process parameters to obtain the most effective parameters of extrusion-cooking process for foamed starch-based materials.

2 Materials and methods

2.1 Materials and mixtures preparation

During tests four types of starches (different botanical origin) were used: potato, wheat, corn and chemically modified corn starch. Various functional additives were used: 99.5% pure pharmaceutical glycerol, Plastron foam PDE, poly(vinyl) alcohol, guar gum, xanthan gum, food gelatine, baking soda, albumin, carrageenan, E471 monoglyceride, technical talc. These additives are used as foaming, emulsifying, stabilizing products [39-41]. They are used in plastic production and also used in bakery, confectionery and other fields of the food industry. Glycerin is a popular plasticizer used to produce TPS starch granules, which are a half-products to obtain biodegradable packaging materials.

Pharmaceutical glycerol was used in an amount from 5 to 25%. The remaining functional additives were used in an amount of 0.5 to 10% by weight of the total raw material mixture. A humidity range 15-20% of processed blends was used.

In the first stage, the starch with functional additives was mixed using a premix mixer produced by Rowag (Rogoźno, Poland). The each mixture has weighed of 5 kg (Figure 1). In order to homogenize all blends, the samples were kept in closed plastic bags for 24 hours in laboratory rooms. Before the extrusion-cooking process, all samples were mixed again.



Fig. 1. Prepared mixtures.

2.2 Extrusion-cooking process parameters

The extrusion-cooking process was carried out using a single screw extrusion-cooker TS-45 (produced by Z.M.Ch. Metalchem, Gliwice, Poland) with L/D = 12 (Figure 2). The extruder was equipped with a water cooling system and temperature sensors for each section, engine load and screw rotation speed.



Fig. 2. The single screw extruder-cooker TS-45 (Z.M.Ch. Metalchem, Gliwice, Poland).

The screw rotational speed was set at 100 and 130 rpm, the temperature profile along the barrel sections (from the feeding section (I) to the head) was relative to type of the starches and varied from 80 up to 140°C (Table 1).

The product was cut using a high-speed knife mounted at the die. The speed was adjusted to the dimensions of the extrudates.

The extrusion-cooking process and the various parameters were continuously monitored and recorded.

Table 1. The temperature profile along the barrel sections.

Barrel section	Blends based on potato starch	Blends based on wheat starch	Blends based on corn starch and corn starch chemically modified
I	80	80	80
II	120	160	140
Head	80	120	100

Due to the specificity and intensity of expansion process, the size of the die was individually selected based on the operator's experience. The aim was to achieve optimum and stable process parameters. In the first stage three different diameters of the die were used: 3, 4 and 5 mm.

Moreover, circular die with the internal diameter of 5 mm was selected to obtain annular cross-section samples. After the extrusion-cooking, the foams were cooled down at room temperature and dried in an air oven at 40°C for 24h (Fig. 3).



Fig. 3. Air oven.

Various mixtures, humidity, temperature ranges, die size were used to see how the extrusion-cooking process and the properties of starch-based foamed packaging materials are changed.

3 Results and Discussion

Preliminary tests have shown that potato starch is susceptible to dextrinisation in higher temperature (over 120°C). It caused sticking and burning of the extrudates (Figure 4). So a decision was made to process potato starch at temperatures below 120°C.



Fig. 4. Dextrinisation of starch.

The results of the first stage also showed that the use only a two additives allowed to obtain satisfactory results in starch-based foamed products processing. Ingredients such as glycerin, guar gum, xanthan gum, food gelatin, baking soda, albumin, carrageenan monoglyceride E 471 and technical talc did not provide the expected results to obtain right form of foamed materials at established process configuration.

Materials produced with these substances were characterized by a compact structure, a small, uneven shape and lack of pores. In addition the extrusion-cooking process was disrupted - sticking of the screw and cylinder, uneven flow of raw material, disturbance of the process expansion. Based on the obtained results, a decision was made to reject most of these additives and use only two components in the further study: Plastronfoam PDE (Figure 5) and poly(vinyl) alcohol (Figure 6).



Fig. 5. Starch-based foam materials with Plastronfoam PDE addition.



Fig. 6. Starch-based foam materials with poly(vinyl) alcohol addition.

Plastronfoam PDE foaming agent is used for foaming thermoplastic elastomers and PVC. The supplier of this ingredient was VGT Polska Sp.zo.o. from Cracow, Poland. Based on the analysis of the first stage and product card, it was decided to use a PDE foaming agent in an amount from 1 to 3% in the mixture. The addition of PDE more than 3% affected negatively on the quality of the extrudates and the extrusion-cooking process. The basic properties of the Plastronfoam PDE are presented in Table 2.

Table 2. Basic properties of the Plastronfoam PDE foaming agent used in the extruded foam production process (data based on producer information card)

Characteristic	The result
Form	White powder
Active substances	Multi-component system
Content of active substances [%]	100
Beginning of decomposition [°C]	>140
Net price for 1 kg [€]	4.5

Poly(vinyl) alcohol is widely used in the production of biopolymers. The supplier of this ingredient was POCH S.A. from Lublin. As in the case of PDE, the proportion of this component in selected blends was from 1 to 3%. The basic features of poly(vinyl) alcohol are presented in Table 3.

Table 3. Basic properties of the poly(vinyl) alcohol used in the extruded foam production process (data based on producer information card).

Characteristic	The result
Form	White color granules
Smell	Without smell
Density [g cm ⁻³]	1.2 – 1.3
Melting temperature [°C]	160 – 200
Net price for 1 kg [€]	45

During the first stage the influence of the humidity content in the mixture on the extrusion process and the quality of the extrudates were observed. It was noted that the humidity less than 17% and over 19% significantly have a significant impact on the extrusion-cooking process. This made the material more difficult to process by the extruder-cooker, the screw was wrapped at feeding section which finally led to stop the process. Such humidity values not allow to obtain good foamed extrudates. So there was decision to use three different humidity levels: 17, 18 and 19%.

Reducing of the die diameter allowed to check how compression ratio of the process and expansion ratio of the product will changed. Using the smallest die diameter allowed to obtain the best expansion ratio of the foamed starch-based materials. So there was decision to use a die with a diameter $\phi = 3$ mm and a ring die. These kind of products shape is one of the most popular on the market.

The higher screw speed (130 rpm) allowed to obtain products with higher expansion ratio. The extrusion-cooking process was also milder and more effective.

Practically most of the starch types used in the tests allowed to produce foamed starch-based materials using the Plastronfoam PDE and the poly(vinyl) alcohol (Figure 7).



Fig. 7. Foamed starch-based materials with poly(vinyl) alcohol addition, from the left: wheat, corn and potato starch.

The exception was chemically modified corn starch, which was not processed during the trials. Finally, based on the price of basic starch raw material (Table 4), potato starch was recommended for further research.

Table 4. Price for 1 kg each starches which were used during tests.

Starch	Price (€) for 1 kg
potato	0.6
wheat	0.9
corn	0.8
corn chemically modified	2.2

4 Conclusions

Preliminary studies of the extrusion-cooking process of different mixtures allowed to select the best main raw materials and functional components. The most effective process parameters was obtain too. The attention was also paid to the properties of the final product and possible lower cost of raw materials.

Based on the results and observations, the following conclusions were made:

1. A single-screw extruder-cooker type TS-45 can be used for the production of starch-based foamed materials.
2. Different types of starch were used during the study and most of them allow to obtain foamed materials.
3. Extrusion-cooking of potato starch at temperatures higher than 120°C resulted dextrinisation of the starch.
4. Based on the results the most functional additives were rejected and only two components were used in the further study: Plastronfoam PDE foaming agent and poly(vinyl) alcohol.
5. Foaming process of the extrudates was achieved with a small addition of the synthetic additives. Both of them: the Plastronfoam PDE foaming agent and the poly(vinyl) alcohol are the substances which can have a contact with food.
6. Humidity content of the mixtures has a significant impact on the extrusion-cooking process and on the quality of the foamed extrudates.
7. Using the smallest die diameter allowed to obtain the extrudates with the best expansion ratio. In addition, the foaming material was obtained using a ring die.
8. The results of the research have made it possible to obtain starch-based foamed materials which can be tested to check basic properties and compare them with traditional polymeric materials.

5 References

1. Y. U. Nabar, D. Drayback, R. Narayan, *J. Appl. Polym. Sci.* **102**,1(2006)
2. Y. Nabar, R. Narayan, *J. Appl. Polym. Sci.* **101**, 6 (2006)
3. Z. Yang, D. Graiver, R. Narayan, *Polym. Eng. Sci.* **53**, 4 (2013)
4. J. Zhou, J. Song, R. Parker, *Carbohydr. Polym.* **63**, 4 (2006)
5. P. D. Tatarka, R. L. Cunningham, *J. Appl. Polym. Sci.* **67**, 7(1998)
6. J. L. Willett, R. L. Shogren, *Polymer* **43**, 22(2002)
7. C. Bastioli, *Handbook of Biodegradable Polymers*(Rapra Technology Limited, UK, 2005)
8. P. Cinelli, E. Chiellini, J. W. Lawton, S. H. Imam, *Polym. Degrad. Stabil.* **91**, 5 (2006)
9. H. A. Pushpadass, S. G. Suresh Babu, R. W. Weber, M. A. Hanna, *Packag. Technol. Sci.* **21**, 3(2008)
10. Z. Yang, D. Graiver, R. Narayan, *Polym. Eng. Sci.* **53**, 4 (2013)
11. L. Wang, G. M. Ganjyal, D. D. Jones, C. L. Weller, M. A. Hanna, *Adv. Polym. Tech.* **24**, 1 (2005)
12. J. W. Lawton, R. L. Shogren, K. F. Tiefenbacher, *Ind. Crop. Prod.* **19**, 1 (2004)
13. J. F. Zhang, X. Sun, *J. Appl. Polym. Sci.* **106** (2007)
14. M. Mitrus, L. Mościcki, *Chemical Engineering Research and Design* **92** (2014)
15. A. L. Chaudhary, M. Miler, P. J. Torley, P. A. Sopade, P. J. Halley, *Carbohydr. Polym.* **74**, 4(2008)
16. El-Sonbati, *Thermoplastic Elastomers* (InTech, Croatia, 2012)
17. P. R. Salgado, V. C. Schmidt, S. E. Molina Ortiz, A. N. Mauri, J. B. Laurindo, *J Food Eng.* **85**, 3 (2008)
18. R. L. Shogren, J. W. Lawton, K. F. Tiefenbacher, *Ind. Crop. Prod.* **16**, 1(2002)
19. Y. X. Xu, Y. Dzenis, M. A. Hanna, *Ind. Crop. Prod.* **21**, 3 (2005)
20. R. Agbisit, S. Alavi, E. Cheng, T. Herald, A. Trater, *J. Texture Stud.* **38**, 2 (2007)
21. H. Chanvrier, F. Desbois, F. Perotti, C. Salzmann, S. Chassagne, J. C. Gumy, I. Blank, *Carbohydr. Polym.* **98**, 1 (2013)
22. A. Wójtowicz, A. Kolasa, L. Mościcki, *Pol. J. Food Nutr. Sci.* **63**, 4(2013)
23. M. Wu, D. Li, L. J. Wang, N. Özkan, Z. H. Mao, *J. Food Eng.* **98**, 4(2010)
24. A. C. Conti e Silva, R. J. Da Cruz, J. A. Gomes Arêas, *Meat Science* **84** (2010)
25. Ş. İbanoğlu, P. Ainsworth, E. A. Özer, A. Plunkett, *J. Food Eng.* **75** (2006)
26. A. Bouasla, A. Wójtowicz, M. Nasereddine Zidoune, *LWT - Food Science and Technology* **75**(2017)
27. N. D. Frame, *The Technology of Extrusion Cooking*. (Blackie Academic & Professional, New York, USA 1994)
28. R. Guy, *Extrusion Cooking. Technologies and Applications* (Woodhead Publishing Limited, Cambridge, U.K. 2001)
29. A. Altskär, R. Andersson, A. Boldizar, K. Koch, M. Stading, M. Rigdahl, M. Thunwall, *Carbohydr. Polym.* **71**, 4(2008)
30. J. M. Ramos Diaz, S. Kirjoranta, S. Tenitz, P. A. Penttilä, R. Serimaa, A. M. Lampi, K. Jouppila, *J. Cereal Sci.* **58**, 1(2013)
31. L. Averous, N. Boquillon, *Carbohydr. Polym.* **56**, 2(2004)
32. L. P. B. M. Janssen, L. Mościcki, *Thermoplastic Starch* (Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany 2009)
33. H. F. Mark, *Encyclopedia of Polymer Science and Technology* (Concise, Third Edition, Wiley-Interscience, A John Wiley & Sons, Inc., Hoboken, New Jersey, USA 2007)

34. J. Ruiz-Ruiz, A. Martínez-Ayala, S. Drago, R. González, D. Betancur-Ancona, L. Chel- Guerrero, *Food Science of Technology* **41** (2008)
35. T. Oniszczyk, R. Pilawka, *Przem. Chem.* **92**, 2(2013)
36. T. Oniszczyk, A. Wójtowicz, L. Mościcki, M. Mitrus, K. Kupryaniuk, A. Kusz, G. Bartnik, *Int. Agrophys.* **30**, 2 (2016)
37. T. Oniszczyk, M. Mitrus, A. Wójtowicz, L. Mościcki, *Przem. Chem.* **94**, 10 (2015)
38. T. Oniszczyk, R. Pilawka, A. Oniszczyk, *Przem. Chem***92**,8 (2013)
39. A. Wójtowicz, *Polish Journal of Food and Nutrition Sciences* **57**, 3A (2007)
40. A. Rutkowski, S. Gwiazda, K. Dąbrowska: *Kompendium Dodatków do Żywności* (Wydawnictwo Hortimex, Polska, 2003)
41. L. Mościcki, *Extrusion-Cooking Techniques* (Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany2011)