Chemical Composition and Characterization of Thermoplastic Starch Biomicrocomposites and Hybrids, Reinforced with Latex and Cellulose Microparticles

Stavros Drakopoulos[1,2,*], Georgios C. Psarras[1], László Lendvai[2] , Ákos Kmetty[2,3], József Karger-Kocsis[2,3]
[1]Department of Materials Science, Universty of Patras, GR-26504, Patras, Greece
[2]Department of Polymer Engineering, Faculty of Mechanical Engineering, Budapest University of Technology and Economics, Műegyetem rkp. 3., H-1111 Budapest, Hungary
[3]MTA–BME Research Group for Composite Science and Technology, Műegyetem rkp. 3., H-1111 Budapest, Hungary

msci1347@upnet.gr

ABSTRACT SUMMARY:
Thermoplastic starch/latex biocomposites attracted the scientific interest because of their biodegradability and their potential applications in packaging industry. In the present study, biomicrocomposites of thermoplastic starch with latex or/and cellulose microparticles were prepared via twin-screw extruder and compression molder, varying the filler concentration. Mechanical, dielectric and thermal properties were investigated in a wide frequency and temperature range.

INTRODUCTION:
In the last few decades, the scientific interest was focused to biodegradable polymers due to their biobased origin and their environmentally friendly behavior. The most examined biobased polymer is starch, which can be found in potato, corn, rice, pea and other plants. Thermoplastic starch can be developed with the gelatinization of starch granules while shearing and heating.[1,3,5] A major disadvantage of thermoplastic starch is its low mechanical properties. As a result of that, biobased reinforcement fillers are added like polycaprolactone, poly(lactic) acid or cellulose and others.[2-4]
In the present work, we investigated the influence of microcellulose (B600 & UFC100) as a filler in a thermoplastic starch matrix and microcellulose/Latex microparticles in a thermoplastic starch matrix to form hybrid biocomposite materials. We examined the mechanical, dielectrical and thermal properties of those biocomposites.

EXPERIMENTAL METHODS:
Thermoplastic starch /cellulose biomicrocomposites and thermoplastic starch /latex-cellulose hybride biomicrocomposites have been prepared by melt mixing in a twin screw extruder (Labtech Scientific LTE 26-44 twin screw), granulation (Labtech LZ-120/VS) and then further melt mixing in an internal mixer (PLASTI-CORDER). For the specimens manufacturing, a compression molding machine (COLLIN P200 E) was also used. Before the extrusion, the raw materials were stored in a Climacell at 35°C and 50% humidity for 6 days prior to extrusion. After the extrusion, the extruded materials were also stored in the same conditions. After the use of the compression molding machine, the compressed sheets were stored again for exactly 1 week and then the tensile tests were performed.
The tensile tests and morphology investigations were performed via a tensile tester (Zwick Z020) and a Scanning Electron Microscope (JEOL JSM 6380LA).
The B600 highly pure cellulose (ARBOCEL® from J.RETTENMAIER & SÖHNE GMBH +CO) with an average fibre length at 60 μm and average fibre thickness at 20 μm according to the manufacturer. The UFC100 highly pure cellulose (ARBOCEL® UFC from J.RETTENMAIER & SÖHNE GMBH +CO) an average fibre length at 8 μm and average thickness at 2 μm according to the manufacturer. The Latex (Natur Latex dry content 60%, Varicham Ltd, Hungary) with a 60% w/w pure latex and 40% w/w water.
The composites have been separately immersed into twelve different mixtures including the reference material as shown in the table below:

no.	Materials
1	Thermoplastic Starch Reference
2	Thermoplastic Starch + 5% B600
3	Thermoplastic Starch + 10% B600
4	Thermoplastic Starch + 15% B600
5	Thermoplastic Starch + 20% B600
6	Thermoplastic Starch + 10% UFC100
7	Thermoplastic Starch + 10% Latex
8	Thermoplastic Starch + 10% Latex + 5% B600
9	Thermoplastic Starch + 10% Latex + 10% B600
10	Thermoplastic Starch + 10% Latex + 15% B600
11	Thermoplastic Starch + 10% Latex + 20% B600
12	Thermoplastic Starch + 10% Latex + 10% UFC100

RESULTS AND DISCUSSION:

Systematic tensile tests have been performed in all mixtures 1 week after the compression molding. For this period specimens were kept in storage. In *Figure 1(a)*, the Young's modulus is increasing above the 10% latex + 10% B600 because above this concentration, the reinforcement becomes efficient. In *Figure 1(b)*, the Young's modulus is increasing above the 15% B600 for the same reasons. The most probable explanation for this effect is that in small quantities, the filler is not behaving as a reinforcement and as such, the Young's modulus is decreasing. Above a percentage, the Young's modulus is restored and increased. Also a great hybrid effect was observed because of the good adhesion between the matrix and the reinforcement materials.

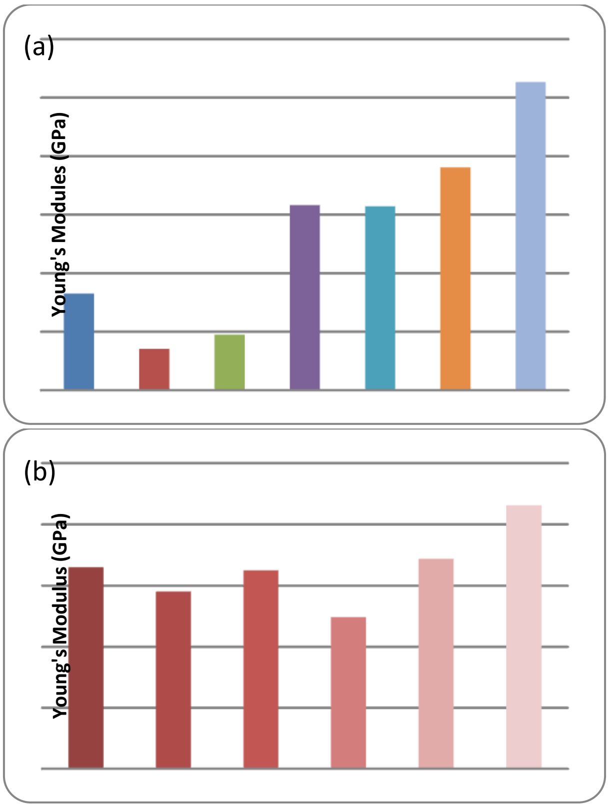

Figure 1. Young's modules for all TPS matrix specimens varying the filler content.
(a) hybrid composite materials, (b) only cellulose containing materials.

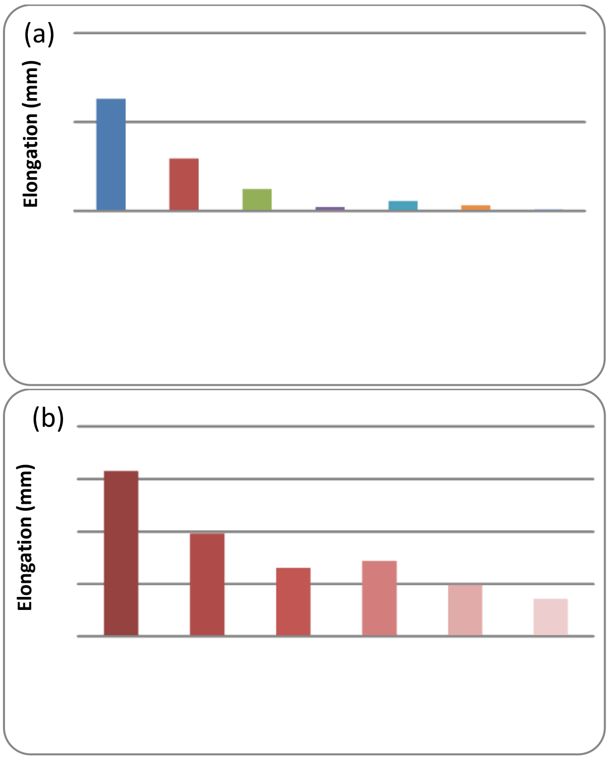

Figure 2. Elongation for all TPS matrix specimens vatying the filler content.
(a) hybrid composite materials, (b) only cellulose containing materials.

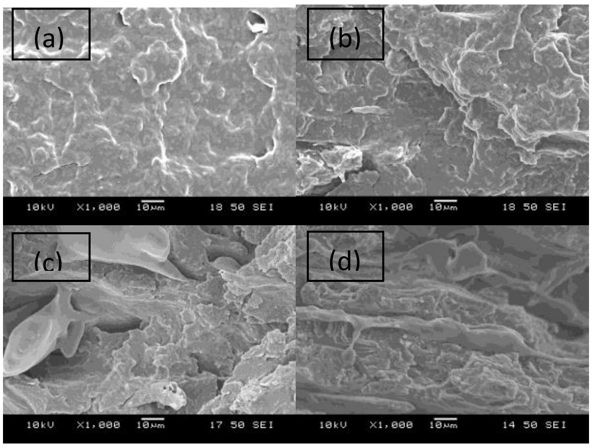

Figure 3. SEM photos for : (a) the TPS Reference material, (b) the TPS+10%B600 material, (c) the TPS+10%Latex+10%B600 material and (d) the TPS+10%Latex+10%UFC100

CONCLUSIONS:

An overall of 12 mixtures have been prepared, including the reference sample. It was observed that the hybrid biomicrocomposite materials exhibit the highest Young's modulus constants. No significant changes have been observed between the reference and the B600-containing composites.

REFERENCES:
[1] T.Tábi, *et al.* Express Polymer Letters, Release 4; 659 (2010)
[2] M.Hietala, *et al.* European Polymer Journal, Release 49; 950 (2013)
[3] S.C.Kiing, *et al.* J Polym Environ, Release 21; 995 (2013)
[4] Y.Zhang, *et al.* J Polym Environ, Release 21;122 (2013)
[5] K.Majdzadeh-Ardakani, *et al*. Carbohydrate Polymers, Release 79; 547 (2010)

ACKNOWLEDGEMENTS:
This research was realized in the frames of TÁMOP 4.2.4. A/1-11-1-2012-0001 „National Excellence Program – Elaborating and operating an inland student and researcher personal support system". The project was subsidized by the European Union and co-financed by the European Social Fund. The research was supported by the Hungarian Scientific Research Fund (OTKA K109409). This work is connected to the scientific program of the „Development of quality-oriented and harmonized R+D+I strategy and functional model at BME" project. This project is supported by the New Széchenyi Plan (Project ID: TÁMOP-4.2.1/B-09/1/KMR-2010-0002).