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Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

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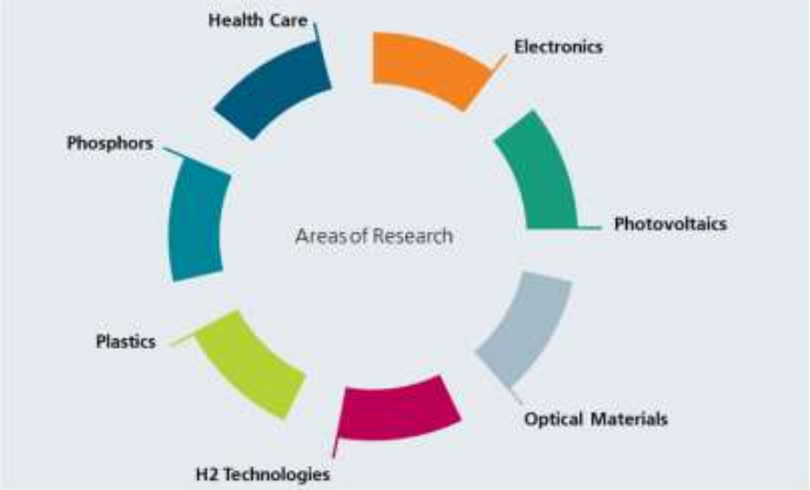
Outline

Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

1. Introduction: Fraunhofer IMWS

The Fraunhofer IMWS at a glance

Methodologically oriented cross-sectional institute for microstructure diagnostics and microstructure design.



- Founded in 1992, since 2016 independent institute
- Locations in Halle (Saale), Schkopau and Soest
- Institute Director Erica Lilleodden
- Budget 27.41 million euros
- Industrial yield 24.28 %
- 352 employees

Non-profit association for application-oriented research and development for the benefit of economy and society.

- **President Prof. Holger Hanselka**
- **Founded 1949**
- **Leading applied research in Europe**
- **Innovation with utility value for industry and society**
- **76 institutes and facilities**
- **Headquarters Munich**
- **30.800 employees**
- **Budget EUR 3 billion**

As of: June 2023

Scientific institutions and facilities at the Weinberg Campus

- Fraunhofer Institute for Microstructure of Materials and Systems IMWS
- Fraunhofer Center for Silicon Photovoltaics CSP
- Fraunhofer Institute for Cell Therapy and Immunology IZI
- Martin Luther University Halle-Wittenberg MLU
- Helmholtz Centre for Environmental Research GmbH
- Leibniz Institute of Agricultural Development in Transition Economies (IAMO)
- Max Planck Institute for Microstructure Physics μΦ
- Leibniz Institute for Plant Biochemistry IPB
- Technologie- und Gründerzentrum Halle GmbH TGZ

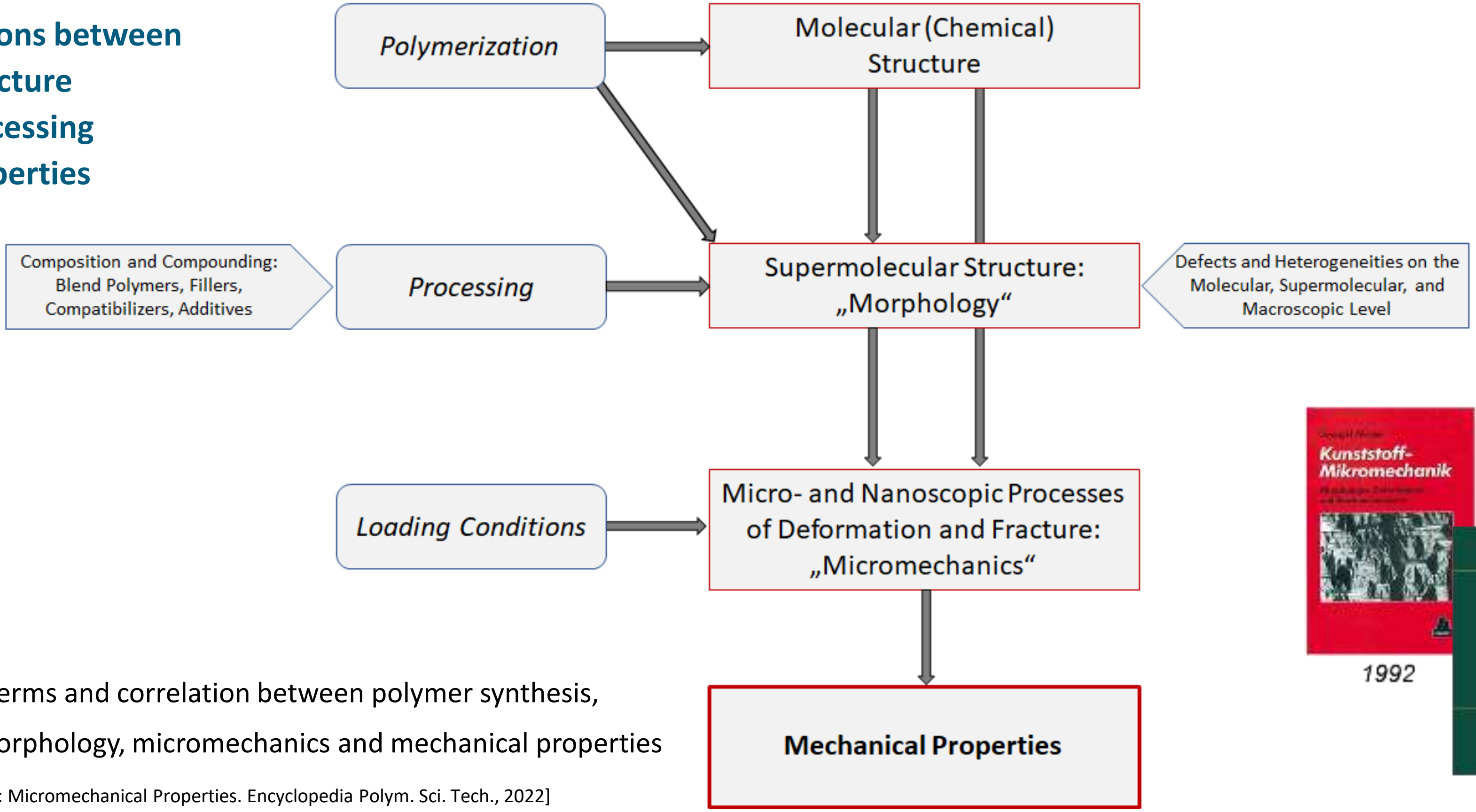
Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

1. Introduction: Morphology and micromechanics of polymers

Morphology and micromechanics: 50 years of research in Halle, Schkopau, and Merseburg

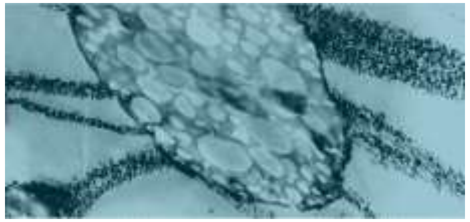
Correlations between

- **structure**
- **processing**
- **properties**



Definition of terms and correlation between polymer synthesis, processing, morphology, micromechanics and mechanical properties

[M. Slouf, S. Henning: Micromechanical Properties. Encyclopedia Polym. Sci. Tech., 2022]



Goerg H. Michler
Morphology and Micromechanics of Polymers

1992

2016

Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

1. Introduction: The conversion problem

The chemical industry in the middle of Germany: Conversion from coal, oil and gas to biobased feedstock

challenges
- **economic crisis**
- **climate change**
- **ecological impact of polymer waste**

→

solutions
- **political decisions**
- **national economic support (money)**
- **technology push**

„Leuna-Werke“ (since 1916)
„BuNa-Werke“ (since 1936)
first synthetic rubber BR

brown coal open-cast mining in Germany

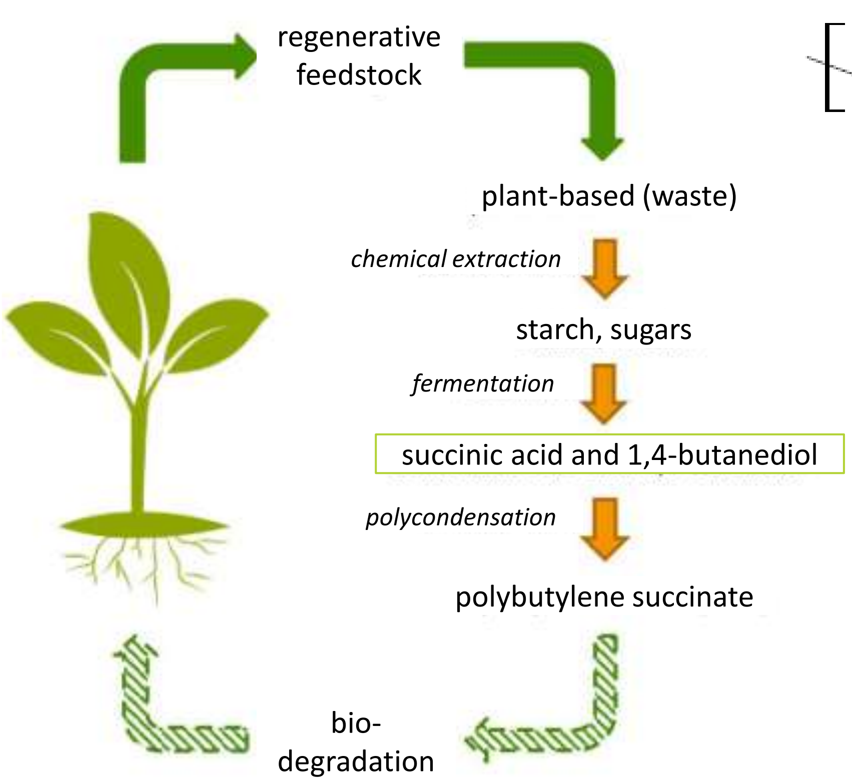
petrochemistry (since 1960)

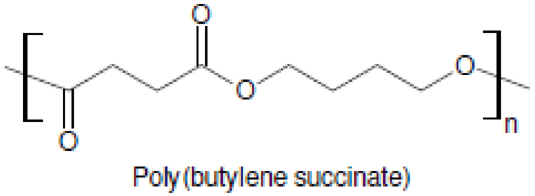
„Bioökonomieregion Mitteldeutschland“
bio-economy cluster
www.mitteldeutschland.com/de/biooekonomie/

2. Polybutylene Succinate (PBS)

Green synthesis and applications of PBS

Sustainable products made of polymers
biobased, recyclable and biodegradable polybutylene succinate PBS



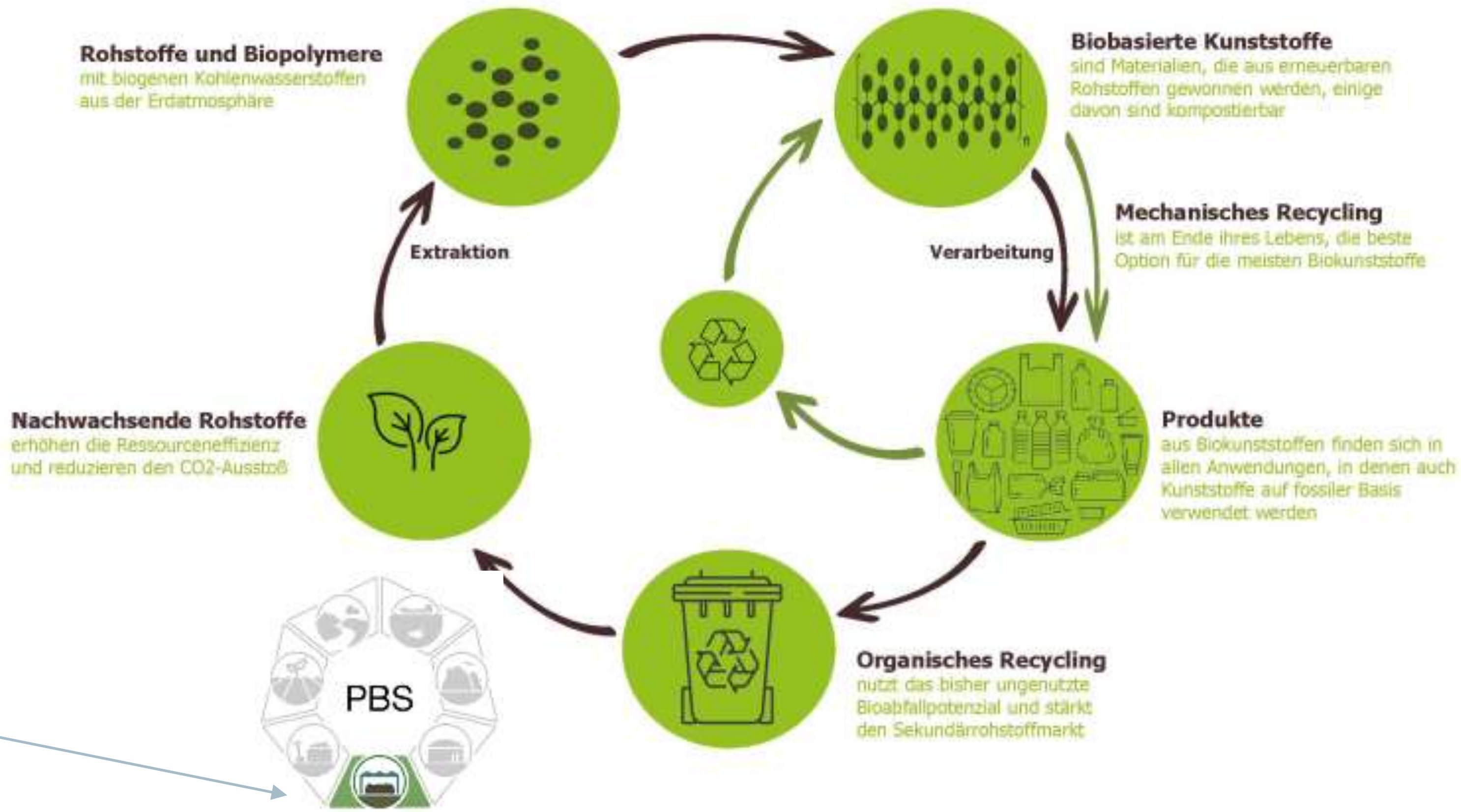


2. Polybutylene Succinate (PBS)

Green synthesis and applications of PBS

Sustainable products made of polymers
biobased, recyclable and biodegradable polybutylene succinate PBS

- Substitution of commodity plastics based on fossile feedstocks (PP, PE, PS, …) by a biobased polymer from **plant waste**
- **Development of products for demanding applications, e.g., automotive (car interior and exterior parts)**
- **Design for recycling**: Supporting circular economy of polymers
- **Compostable** at the end of life: PBS decomposes into water and CO_2 through naturally occurring degrading enzymes and microorganisms (under industrial composting conditions)



Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

3. Direct compounding injection moulding (DCIM)

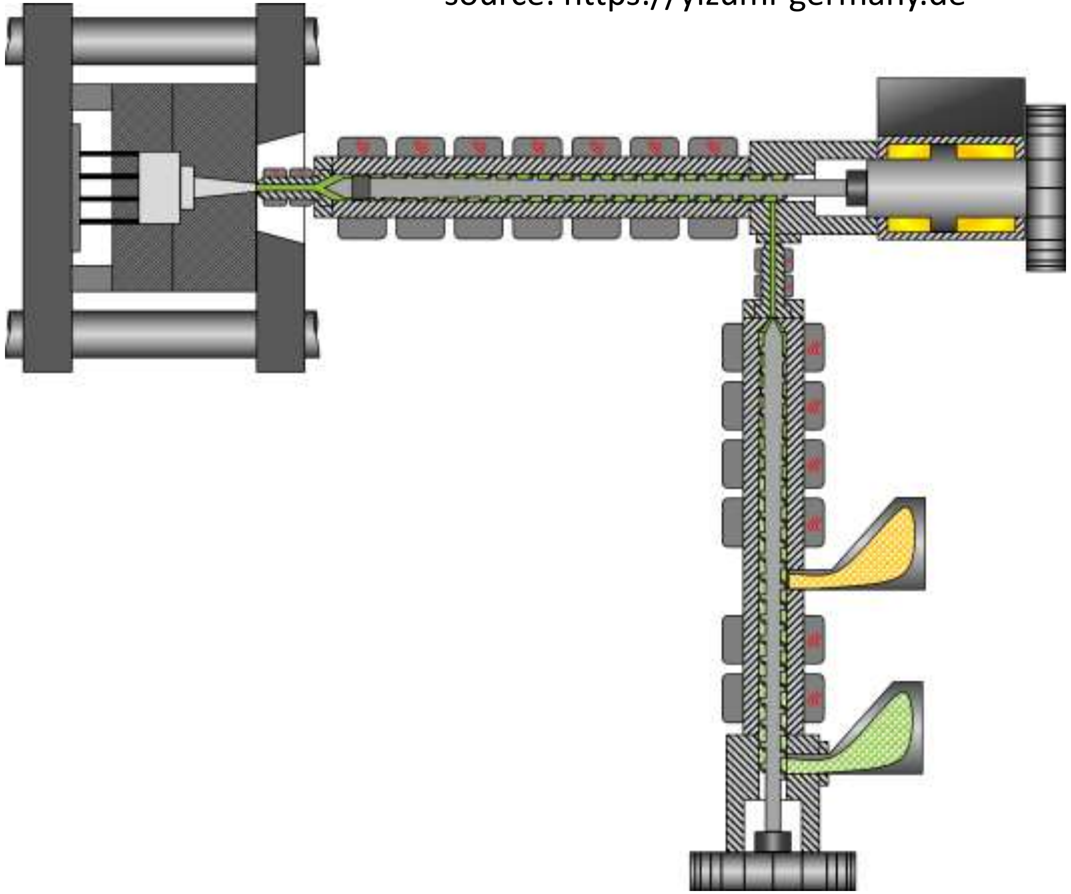
Design and advantages

CO_2 reduction in the value chain

Ideal for biopolymers, bio-based and bio-degradable

Perfect for circular economy and recycling

source: https://yizumi-germany.de



Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

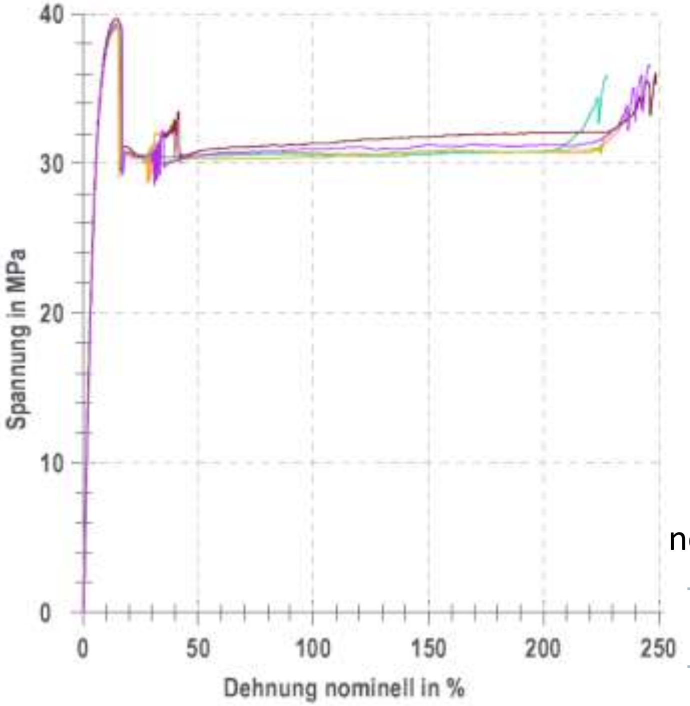
4. PBS for injection moulding applications: Project and first results

Mechanical properties and molecular weight

Benchmark: toughened PP for automotive appl. MFR > 7g/10 min
impact strength (RT) > 15 kJ/m²

Sample Materials: PBS 1 BioElan A005 MFR 5 g/10 min | PBS 2 BioCelain A140S MFR 40 g/10 min + 10 % layered silicate

PBS 1 BioElan A005



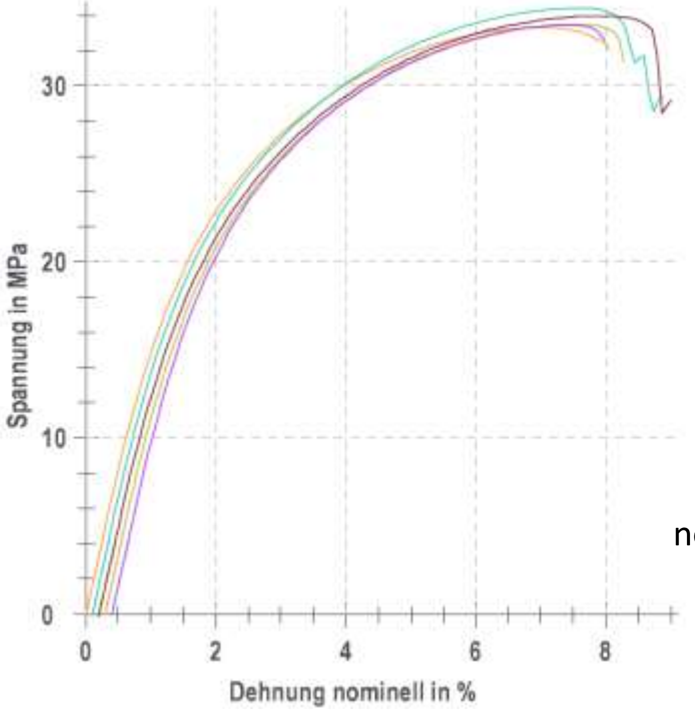
uniaxial tensile test

E_t [MPa]	σ_m [MPa]	ε_b [%]
745	39.3	237

notched impact strength (Charpy)

a_{cN} [kJ/m²]	8.66

PBS 2 BioCelain A140S + 10 % layered silicate



uniaxial tensile test

E_t [MPa]	σ_m [MPa]	ε_b [%]
1633	33.7	8.24

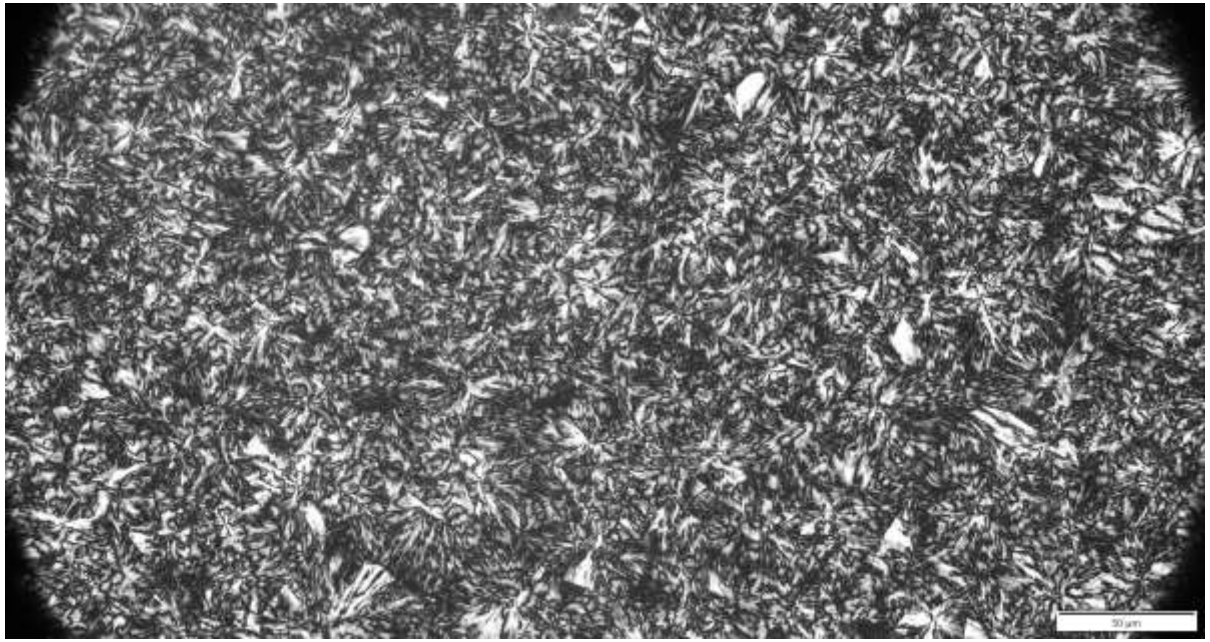
notched impact strength (Charpy)

a_{cN} [kJ/m²]	3.61

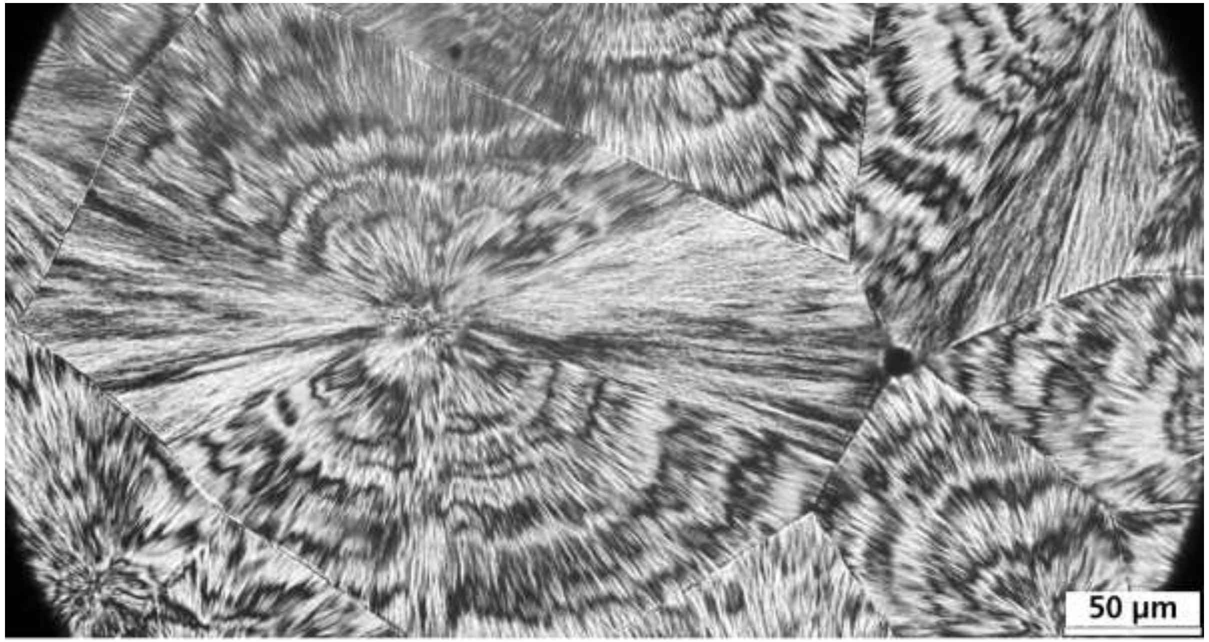
4. PBS for injection moulding applications: Project and first results

Semicrystalline morphology and crystallization control: Shperulite size as a function of crystallization temperature

interspherulitic defects!



solution-cast film of **PBS 1**, isothermal crystallization at 75 °C



solution-cast film of **PBS 1**, isothermal crystallization at 90 °C

Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

4. PBS for injection moulding applications: Project and first results

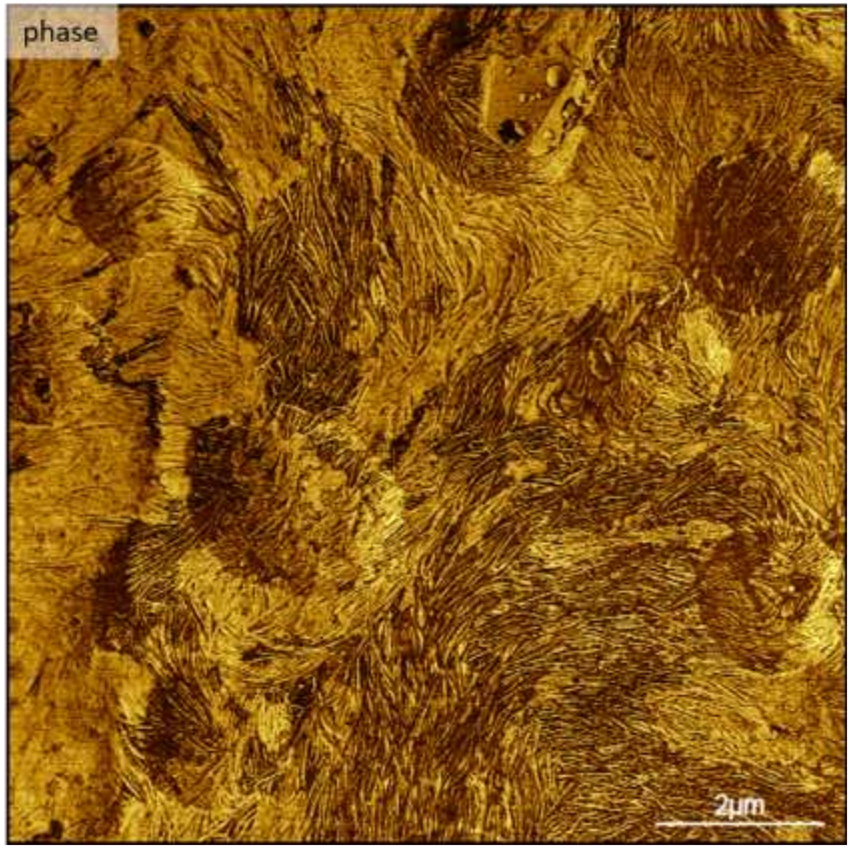
Semicrystalline morphology and crystallization control: AFM images of the lamellar arrangement of spherulites

PBS 1

Bio-Elan A 005 after 75 °C isothermal crystallization

PBS 2

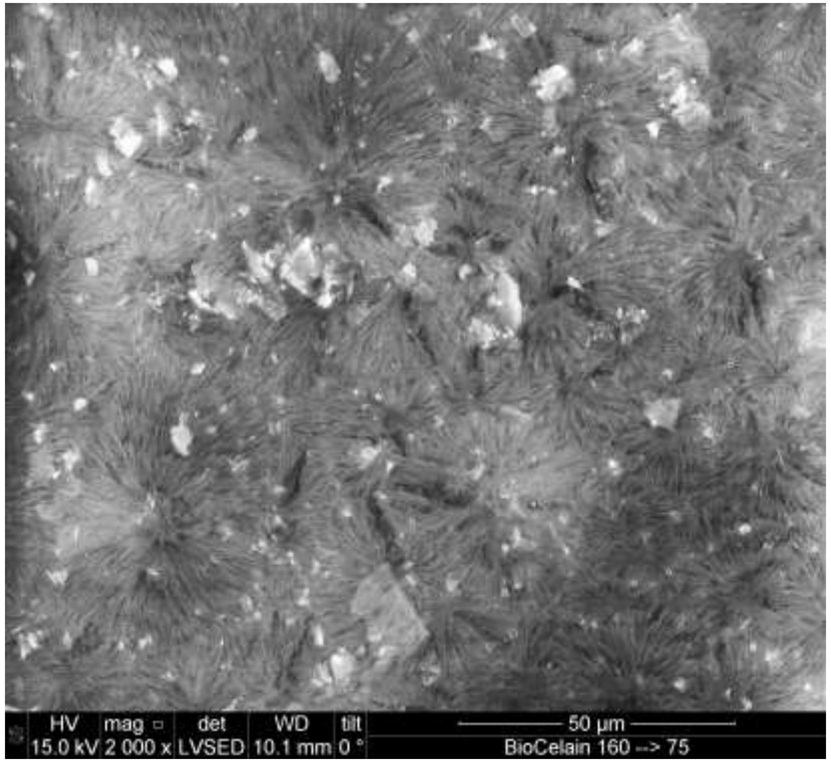
BioCelain A 140 S after 75 °C isothermal crystallization



4. PBS for injection moulding applications: Project and first results

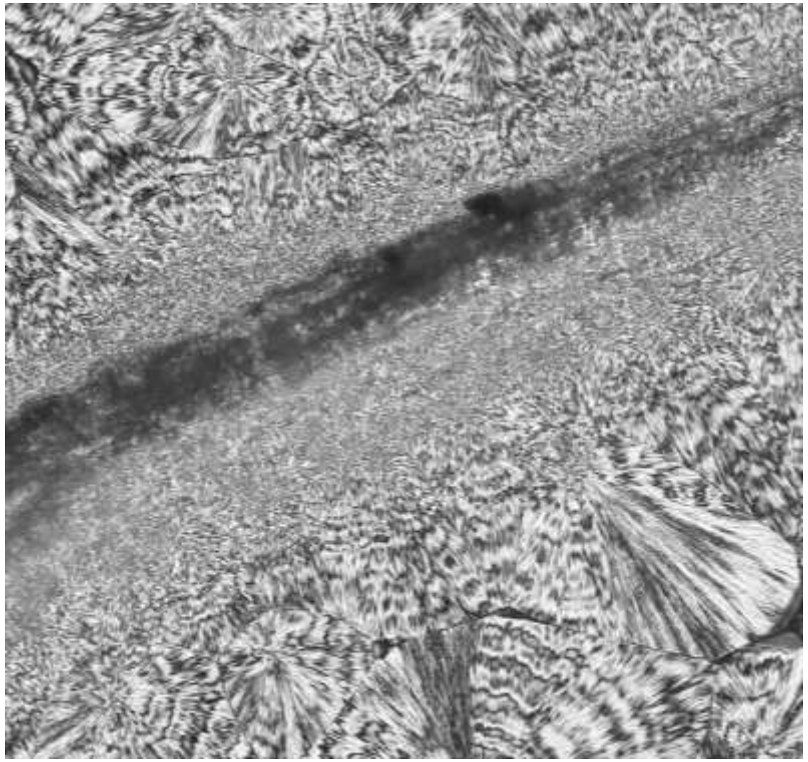
Semicrystalline morphology and crystallization control: Nucleation initiated by fillers?

layered silicates: no nucleation effect



isothermal crystallisation of **PBS 2** with layered silicate at 75 °C: no nucleation

plant fibers (hemp): nucleation effect



isothermal crystallisation of **PBS 1 with hemp fibers** at 90 °C: nucleation of crystal growth on fiber surface

Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

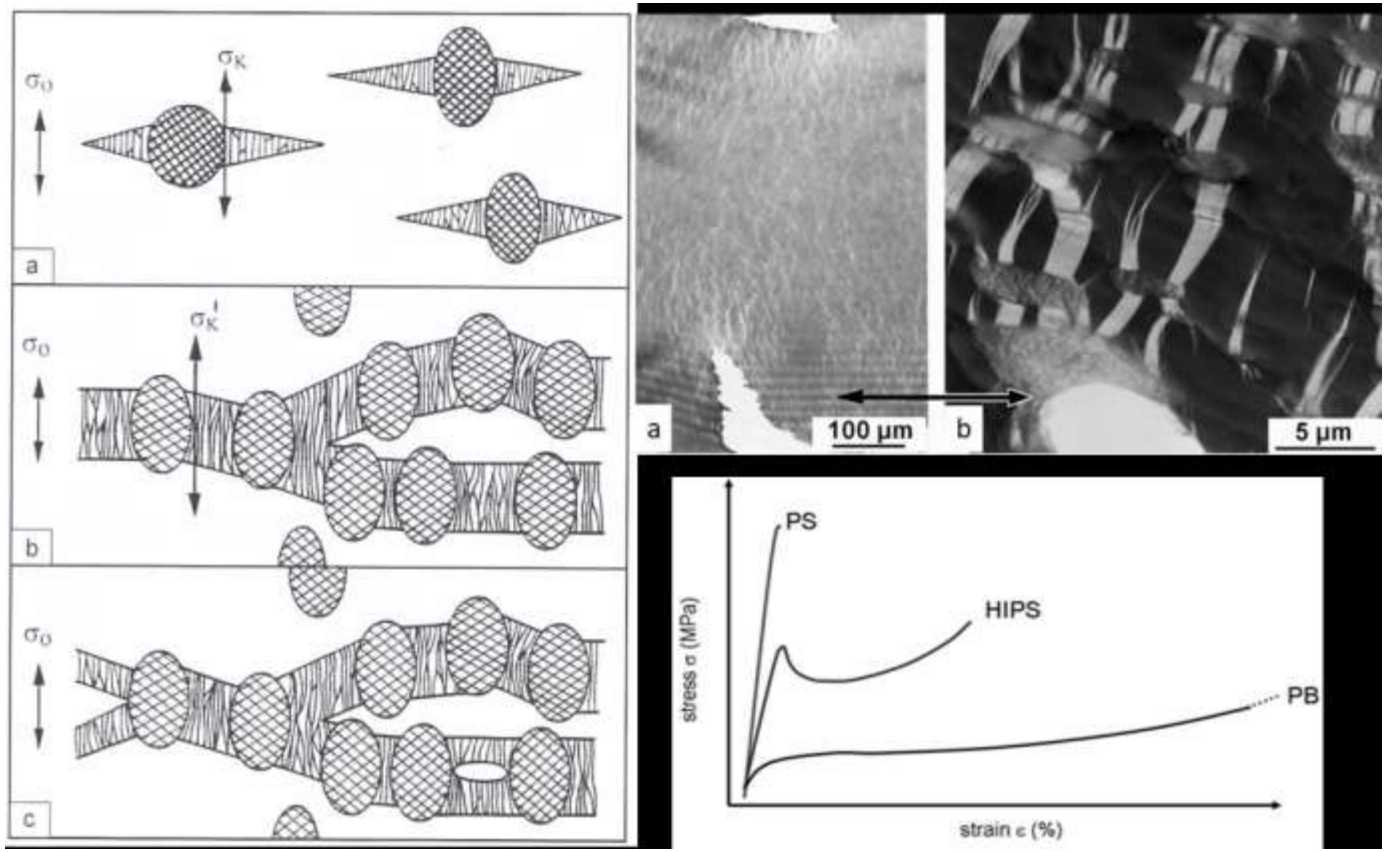
4. PBS for injection moulding applications: Project and first results

The principle of toughness enhancement

Classical mechanism of toughness enhancement shown for high impact polystyrene (HIPS):
a) overview image and b) detail image at higher magnification showing multiple craze initiation in the PS matrix and crack bridging by polybutadiene rubber particles; high voltage TEM image, in situ deformation of a semithin section.

Schematic description of the classical three-stage mechanism in rubber toughened polymers based on multiple crazing, crack bridging and crack stop mechanisms: a) stress concentration σ_K at rubber particles, b) superposition of stress concentration fields with elongation of rubber particles and craze initiation, c) crack bridging and crack stop. after *Michler* (HIPS, TEM images)

[M. Slouf, S. Henning: Micromechanical properties. Encyclopedia Polym. Sci. Tech., 2022, DOI: 10.1002/0471440264.pst199.pub2]



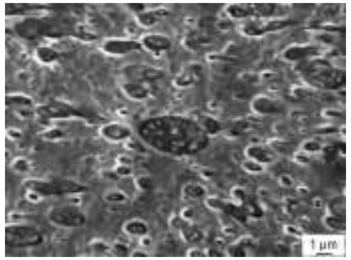
Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

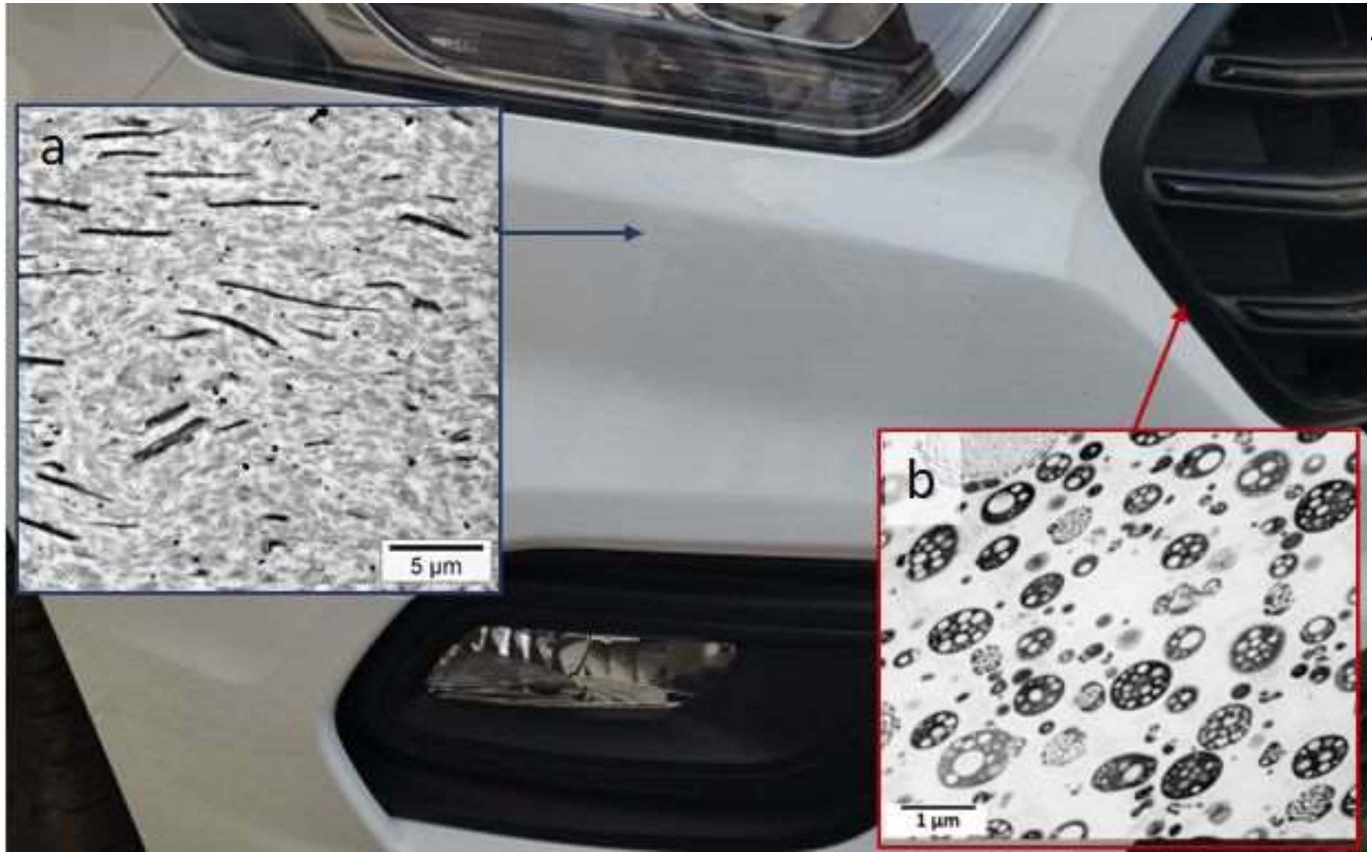
4. PBS for injection moulding applications: Project and first results

The principle of toughness enhancement

PP-EPR + inorganic filler

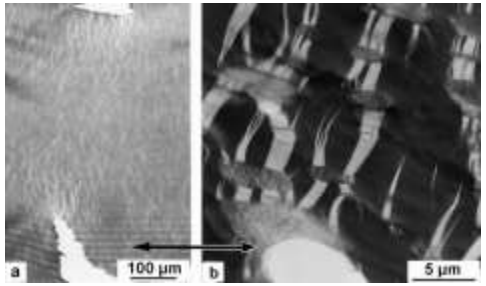
- PP problem: embrittlement at low temperatures (< $T_{g,am}$ approx at – 5 °C) and with low M_w (high MFR)
- heterophasic PP: Introduction of ethylene-propylene-copolymer as a rubbery phase for toughness enhancement





ABS

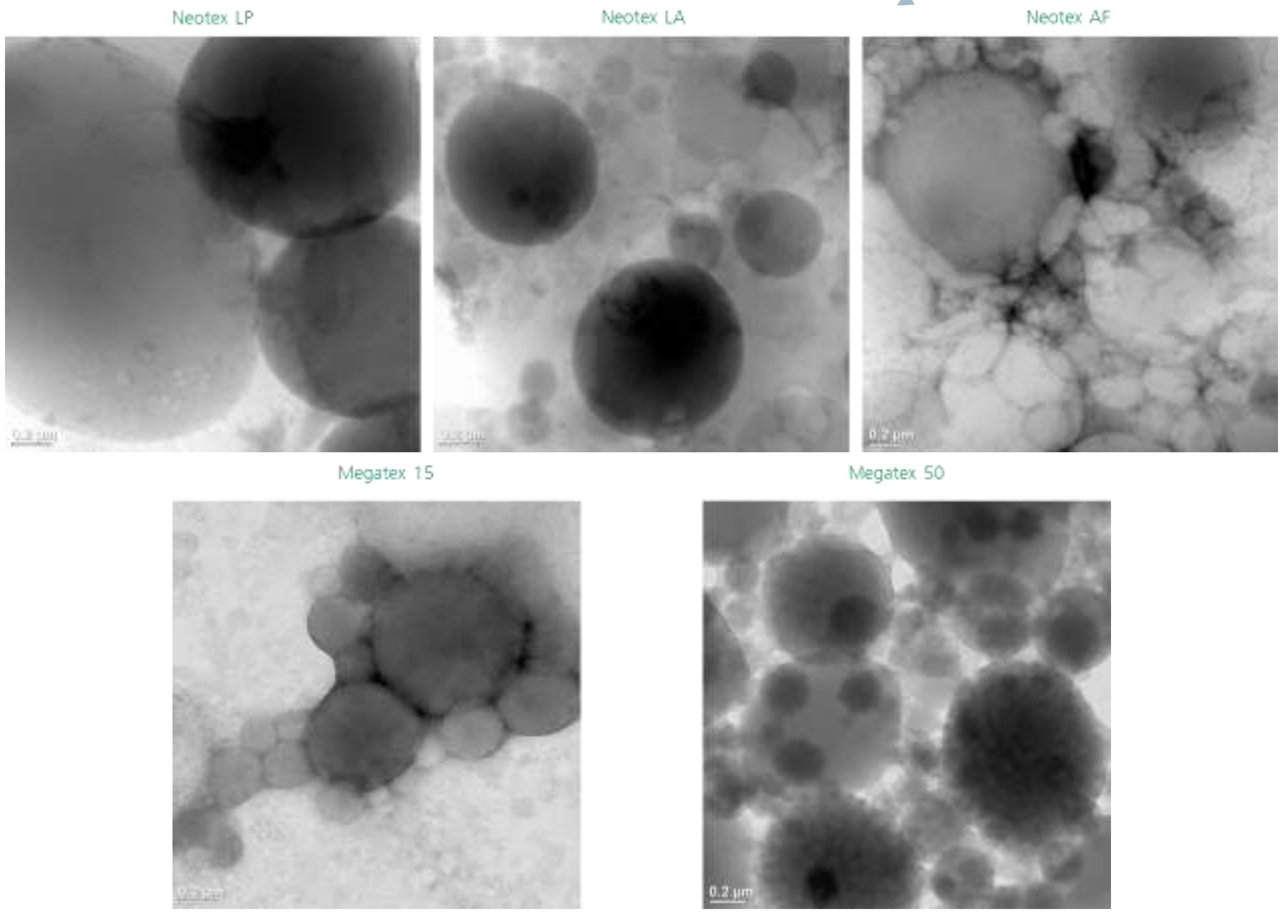
- toughening of the brittle PS matrix by introduction of a rubbery phase (polybutadiene)



Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

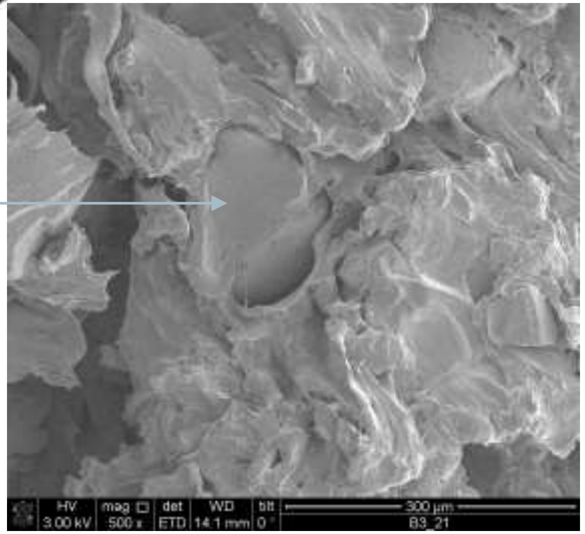
4. PBS for injection moulding applications: Project and first results

Incorporation of rubber modifier: Natural rubber latex particles and recycled tire rubber (ground rubber powder)



rubber particles < 450 µm

not suitable for toughening of brittle polymers



thermoplastic matrix with 60phr ground rubber + compatibilizer

Toughness enhancement of polybutylene succinate (PBS) for injection moulding applications

4. PBS for injection moulding applications: Project and first results

Incorporation of rubber modifier: Natural rubber latex particles

code	description	typical size latex particle size and special features
Neotex LP	natural rubber latex	400 nm to 2,8 µm
Neotex LA	natural rubber latex, low ammonia (0,3 %)	**100 nm to 1 µm**
Neotex AF	natural rubber latex, ammonia-free	**100 nm to 1 µm**
Megatex 15	natural rubber latex, grafted with MMA (15 %)	100 nm to 1,6 µm
Megatex 50	natural rubber latex, grafted with MMA (50 %)	**bimodal distribution with fraction 1: 100 nm to 300 nm, fraction 2: 750 nm to 1,6 µm, some latices with special structure („Salami Particles")**

next process steps:

- cross-linking of latex particles (phys. and chem.)
- spray drying / coagulation
- compatibilization and surface stabilization (layered silicates)
- incorporation of stabilized rubber latex particles in to PBS via DCIM

5. Summary

- PBS is a biobased polymer with a profile of mechanical properties and versatility comparable to PP

- at high MFR (low M_w) essential for efficient injection moulding, PBS suffers embrittlement; the ductile-to brittle transition is going along with a transition from yielding to craze-like deformation mechanisms

- the addition of inorganic filler increases Young´s modulus, but further decreases toughness (as expected)

Toughness enhancement requires

1. Good balance between desired high MFR and appropriate M_w with respect to entanglement M_w of the intercrystalline amorphous phase (tie molecules and entaglements)

2. Crystallization control: Setting the right processing conditions (cooling rate, mould temperature) and introduction of nucleating agents, e.g., (nano- or micro-) cellulose from plant waste

3. Application of classical strategies for toughness enhancement: Incorporation if micro- and nanoscopic rubber particles

Thank you very much for your attention!

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