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Composite materials for the circular economy: Tuning the biodegradability of materials through the use of spent coffee grounds as filler

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Utilization of biomass waste

Our food production generates large amounts of waste biomass, in the shape of all the inedible parts of the plants. While a small percentage is utilized further, e.g. for animal bedding, currently most of it is burned on the fields or put into landfill, generating large amounts of CO_2.

In a zero waste/circular economy approach, we must utilize all parts of the plants to make materials, chemicals and fuels, beside food.

Biomass waste consists mostly of lignocellulose, but, depending on the source, contains additional specific, valuable compounds that can be extracted.

Functional additives from biomass

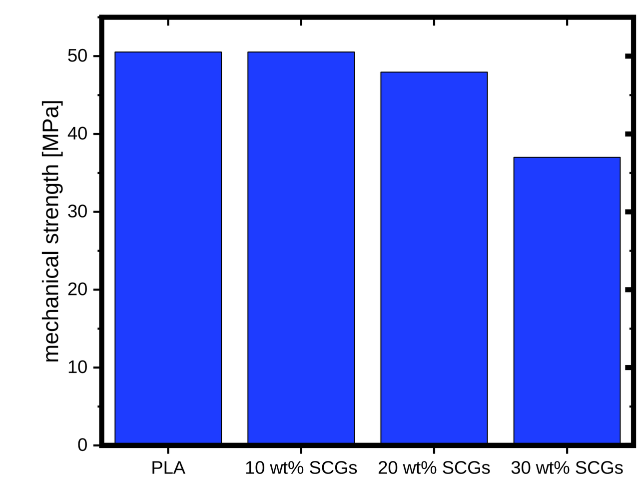
Biomass waste can contain valuable compounds such as antioxidants, proteins or oils, which can be extracted and used as additives.

Spent Coffee Grounds

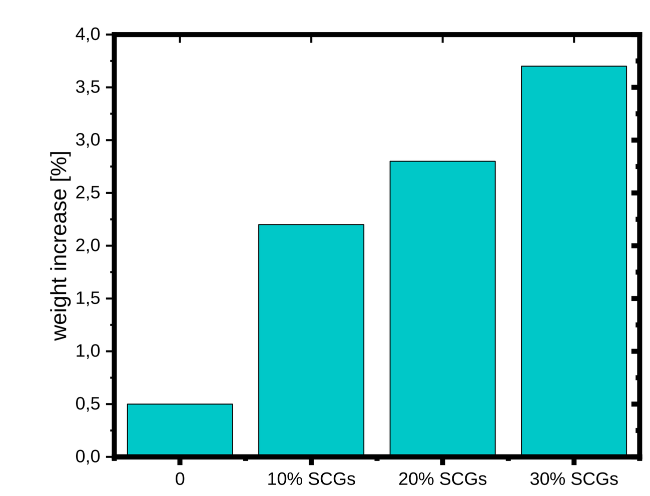
Spent coffee grounds (SCGs) are the leftovers from coffee brewing. They make up a large part of the total waste biomass generated during coffee production (14% of total biomass at harvest / 0.6 kg per kg of coffee beans), and are available around the world. They contain oil, as well as different antioxidants.

PLA – SCGs composites

- Water uptake into the material is increased by the hydrophilic SCGs, leading to faster hydrolysis rates
- Mechanical strength is not affected at lower loadings and can be improved with additives



Mechanical strength



water uptake after 24 h

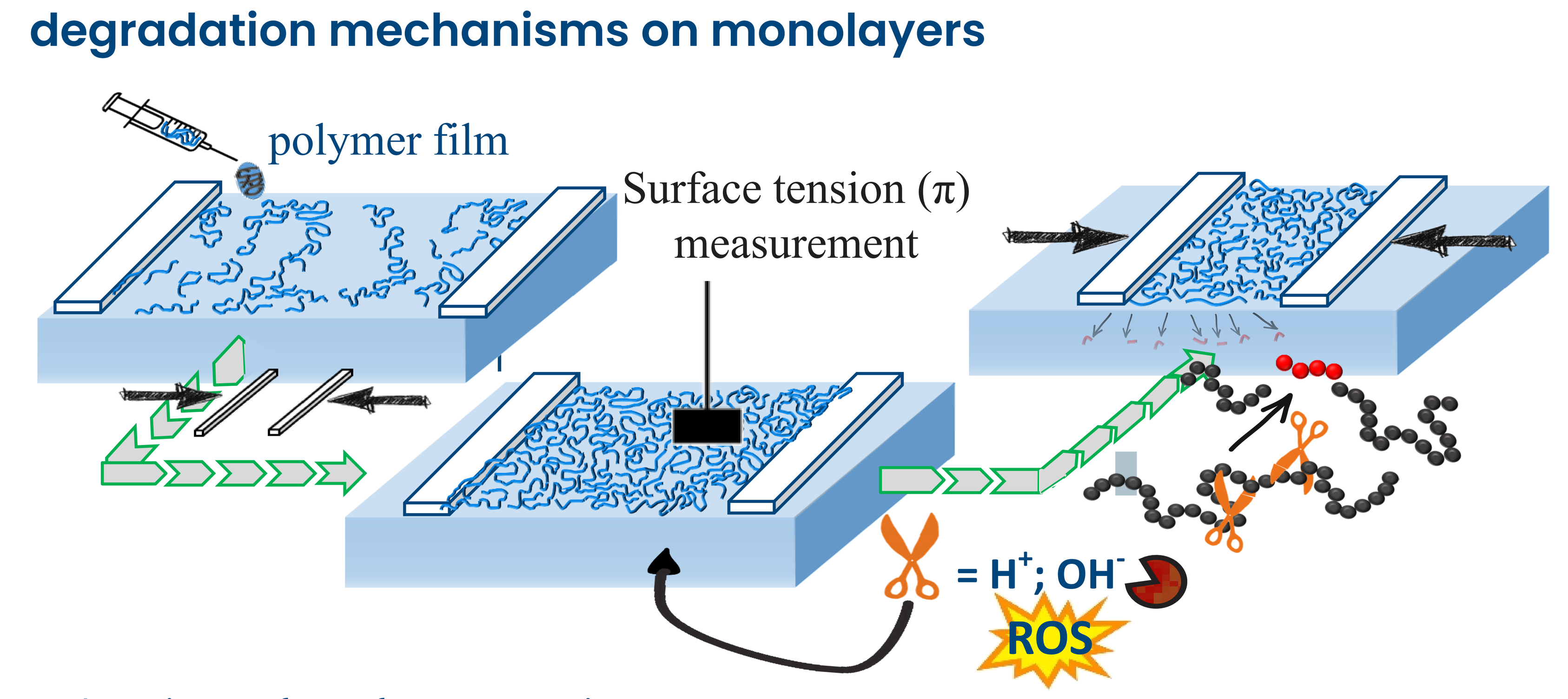
Biodegradability

Biodegradable polymers

Only a few synthetic biodegradable polymers exist so far. They all belong to the group of polyesters and are cleavable by hydrolysis, catalyzed by acid, base or enzymes. Even less of them are also derived from renewable resources:

- Polylactic acid (PLA), derived from sugar via fermentation to lactic acid, industrially compostable
- Polyhydroxyalkanoates (PHAs), a group of polymers synthesized by bacteria such as *P. putida* from different 3-hydroxy carboxylic acids as energy storage, readily biodegradable for short side chains
- Polybutylene succinate (PBS) is only available in 50% bio-based, although it could be fully made from bio-derived 1,4-butanediol. Related polymers such as polybutylene adipate (PBAT) are currently not made from renewable feedstock.

The Langmuir-Blodgett technique for the investigation of polymer degradation mechanisms on monolayers

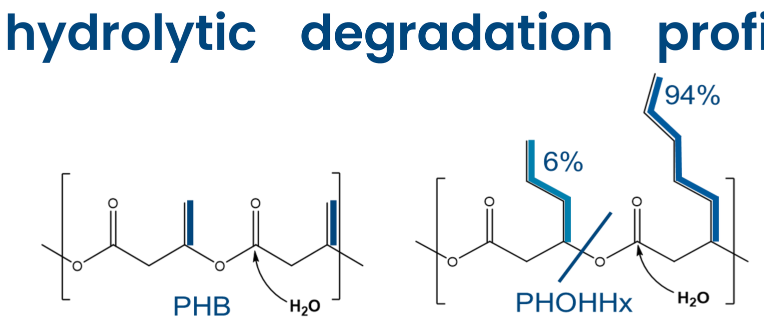


Reduction of surface tension γ = increase in surface pressure π

$\pi = \gamma_{water} - \gamma_{surfactant}$

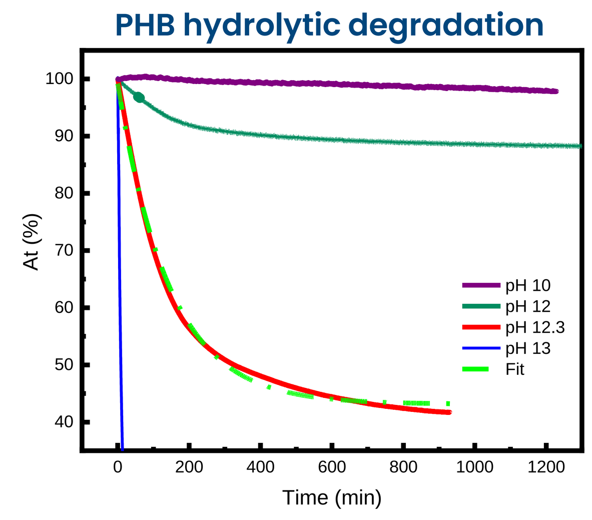
force per unit length

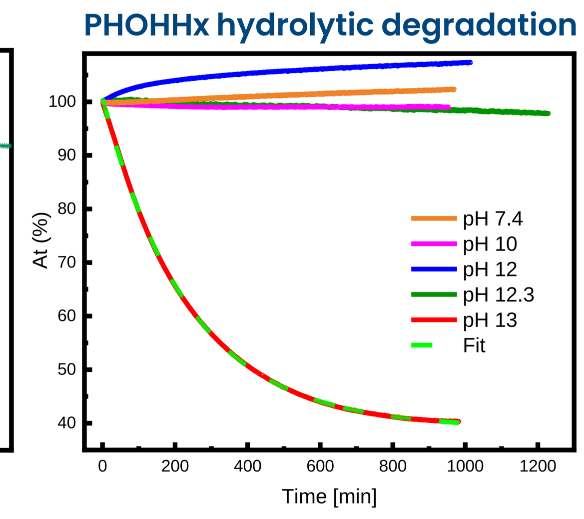
Comparison of hydrolytic degradation profiles of PHB and PHOHHx[a]

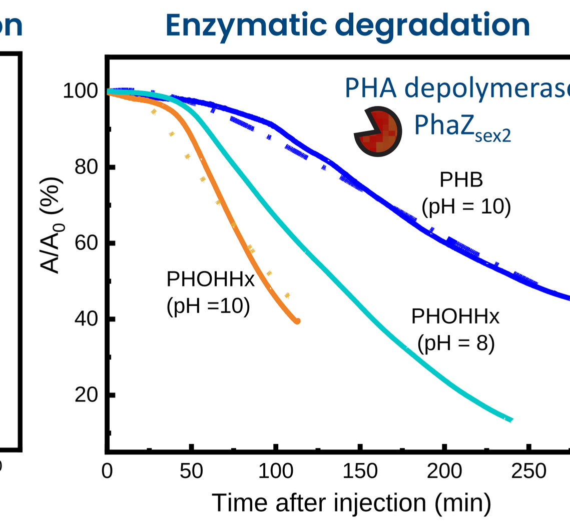


Polyhydroxybutyrate (PHB) and Polyhydroxyoctanoate-co.hexanoate (PHOHHx)

- Amorphous PHB degrades much faster than PHOHHx with increasingly alkaline pH (end cuts)
- The degradation in monolayers is also affected by the solvation of the polymer chains. For PHOHHx with longer, more hydrophobic side chains, the solvation is not as good
- PHA depolymerase can degrade both PHAs at lower pH values (random scission)

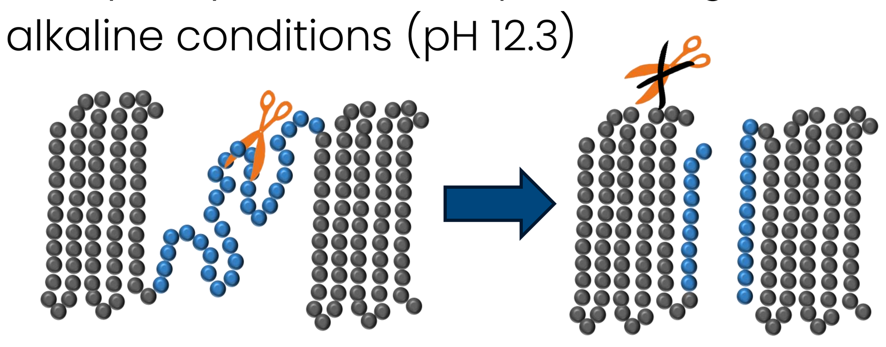




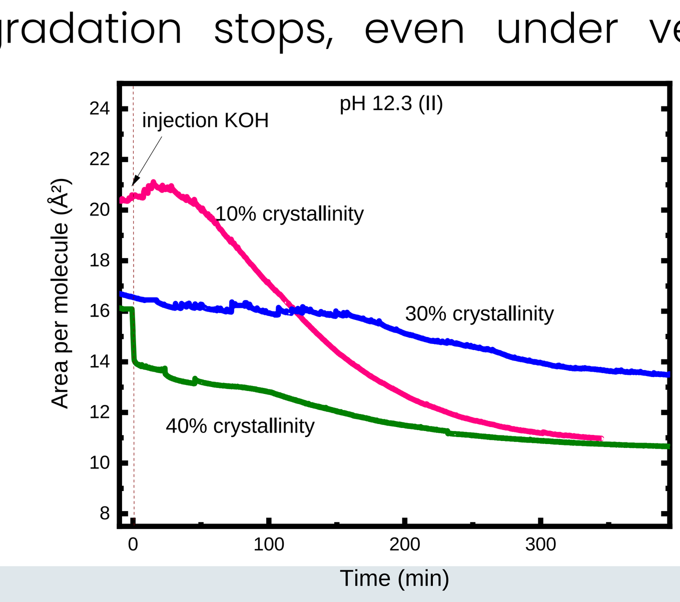


Influence of crystallinity

PHB is degradable up to 30% crystallinity. When chemicrystallization sets in due to hydrolysis of amorphous regions, degradation stops, even under very alkaline conditions (pH 12.3)



- Polymers need to be designed with spacers to avoid this from happening during degradation



Summary

- Addition of biomass waste as filler to biodegradable polymers enables the reduction of costs and material use as well as tuning the degradation behaviour
- The Langmuir-Blodgett monolayer technique can give important insights into the degradation mechanism of polymers and help in the design of new materials
- The degradation profile of PHAs is affected by the length of the side chains and the crystallinity of the polymer

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a) Tarazona N. A. Tarazona, R. Machatschek and A. Lendlein, Biomacromolecules 21 (2), 761-771 (2020)