Biobased Chemistry and Technology
Annual report 2020

WAGENINGEN
UNIVERSITY & RESEARCH

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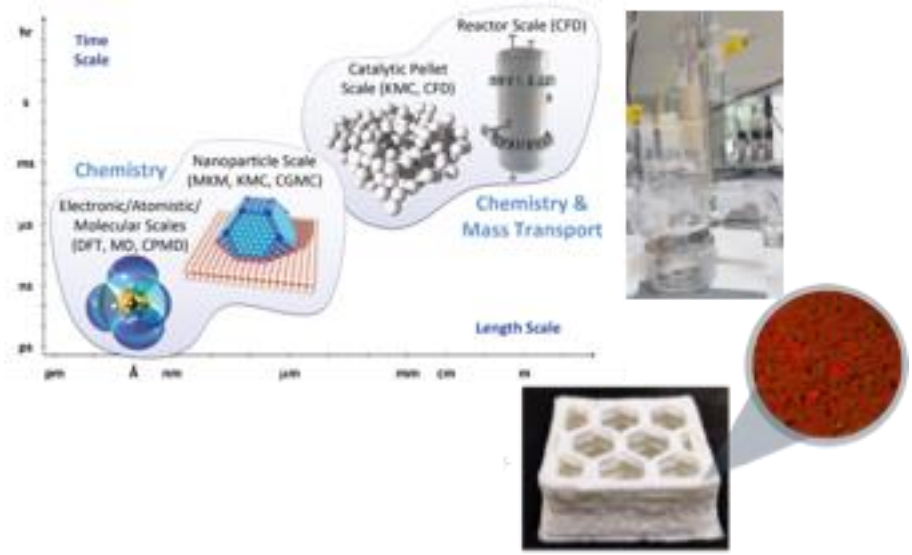
Contents

Introduction

It is my pleasure to present to you the annual report 2020 of the Biobased Chemistry and Technology (BCT) group.
Despite the challenging year due to the Covid crisis the group kept its momentum and even created new momentum with the appointment of Dr. Guanna Li as tenured track assistant professor focussing on computational chemistry and Dr. Akbar Asadi Tashvigh who is expert in modelling and membrane chemistry.
The great momentum of the group was further shown by the promotion of Dr. Costas Nikiforidis to associate professor and the granting of different research proposals in all fields. You will read about these proposals in the next year report.
Unfortunately, Dr. Lars Kiewidt left the group for a career in industry. We are very grateful to him for the years he part of the group; he was a key player in setting up the modelling part of the group and connect it to chemistry.
A very sad moment in 2020 was the passing away of Dr. Piet Buwalda, his presence and input in the carbohydrate chemistry will be dearly missed.



Some highlights/approaches of BCT. Multiscale modeling capabilities (top-left), electrochemical reactor for biofeedstock conversions (top right) and 3D printed structure based on oleosomes/proteins (bottom)

All this made 2020 an interesting and exiting year again and I'm sure you feel the same after reading this report.

With kind regards

Prof. J.H. (Harry) Bitter
Chair holder Biobased Chemistry and Technology

Collaborations

The strategy of the BCT group is to develop fundamental insights in processes relevant for biobased conversions and based thereon suggest improvements of process technology in the biobased economy. This not only requires the incorporation of several length and time scales, but also the knowledge and expertise of multiple disciplines. Therefore, we established collaborations with other groups within and outside Wageningen. Some of our collaborations are summarized in the table below.

Collaborating group in Wageningen	Topic
Environmental Technology (ETE)	Combining chemo and bio-electro-catalysis Modeling of water-energy-material nexus in industrial and urban environments
Organic chemistry (ORC)	Teaching and research proposals
Bionanotechnology (BioNT)	Combining catalysis and NMR in microreactors. Use of natural constructs as carriers for nanoparticles
Physical Chemistry and Soft Matter (PCC)	Education on natural materials
Plant breeding (PBR)	Synergy between plant sciences and biorefinery
Food and Biobased research (FBR)	Different research projects and acquisitions
Bioprocess Engineering (BPE)	Research collaboration
Microbiology (MIB)	Research Collaboration
Food Chemistry (FCH)	Research Collaboration
Food Process Engineering (FPE)	Research Collaboration
Physics and Physical Chemistry of Foods (FPH)	Research Collaboration

The BCT group participates within Wageningen in the research schools VLAG and WIMEK and is part of the Netherlands Institute for Catalysis Research (NIOK) and the Institute for sustainable process technology (ISPT). The group also collaborates intensively with other academic and industrial consortia both within WUR (FBR-Wageningen Research) and outside to address the multi-disciplinary character of the challenges (e.g., within EU projects, TTW, TIFN, ISPT, Center for Biobased Economy (CBBE) and advisory boards such as the advisory board of the VNCI and the bioeconomy federation).
In 2018 Harry Bitter was appointed as adjunct (guest) professor in sustainable catalysis at the department of green chemistry of Monash University in Melbourne Australia. This collaboration will strengthen the biobased activities within Wageningen both at research and education level.

Catalysis/Conversion

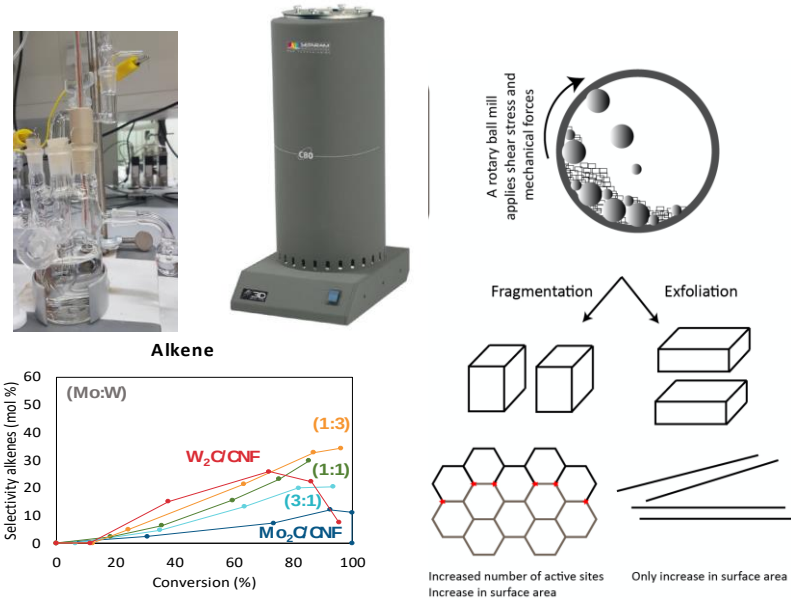
Team Leaders: Dr. Elinor Scott/Prof.dr. Harry Bitter
PhD students/Post doc: Roel Bisselink, Roxani Chatzipanagiotou, Marlene Fuhrer, Xinhua Goerner-Hu, Matthijs van der Ham, Tim Hoogstad, Frits van der Klis, Cynthia Klostermann, Nazila Masoud, Dmitry Pirgach, Edwin Schreuder, Sanne de Smit.
Contact: elinor.scott@wur.nl or harry.bitter@wur.nl


Background and goal

In this theme we develop new catalyst and catalytic conversions routes based on scientific understanding of the catalyst or process. The aim is to develop sustainable conversions routes and processes. To achieve that a multi-disciplinary approach is used i.e. catalyst development goes hand in hand with computational method and multi-scale modelling.
The use of electricity as an energy input to drive the reactions is a part of the theme which grew significantly in the past year.

Main (sub) topics:

Catalyst development for biobased feedstock conversion (heterogeneous, homogenous, bio)
Use of non-noble metal catalyst to replace scarce noble metals
Electrochemical/electrocatalytic conversion of larger (biobased) molecules
Integration of thermal catalysis with electro catalysis/chemistry
CO_2 capture and conversion



Some examples: electrochemical reactor for biobased conversions (top left), commissioning of a C80 calorimeter for operando heat measurement (top middle), modification of carbons by ball milling (right) and steering selectivity of Mo/W-carbide catalysts for deoxygenation of stearic acid (bottom left)

Highlights from last year

-New projects on electrocatalysis (H_2O_2 production (ECCM), conversion of biobased feedstocks) and CO2 capture (NWO-Lift)

-Simone Verhagen, MSc student at BCT, received a scholarship from the Aalt Dijkhuizen fund to continue her reserach around seaweed at Chalmers University of Technology in Gothenburg, Sweden.

Students project envisioned

Thesis subjects are related to the research of the Ph.D. projects described on the next pages. The projects can be either lab based or modelling based. Contact Elinor Scott (Elinor.scott@wur.nl) for further information.

Electrochemical cell design for the production of valeric acid and hydrogen peroxide

Name PhD: Roel Bisselink
Involved staff members: Prof. Dr. J.H. Bitter; Dr. J. van Haveren
Project sponsor: NWO program, TKI BBEG-program, TKI programs
Start/(expected) end date of project: August 2020 – July 2026

Background and goal of project

The energy sector is currently in a transition towards energy generation from renewable sources (wind, water and sun). Moreover, the production of electricity from renewable resources is already cost competitive with its fossil-based counter-part. The (future) availability of cheap electricity therefore presents an opportunity to drive chemical transformations and thus enables electrification / decarbonization of the chemical industry. Possibilities to decarbonize current processes can be found in the electrochemical conversion of 1. bio-based levulinic acid to valeric acid and 2. oxygen to hydrogen peroxide. But to come to an efficient electrochemical process design for these routes it is vital to increase understanding of various fundamental aspects of each conversion.

In previous research electrode materials have been identified enabling efficient conversion of levulinic acid to valeric acid.[1] However, the mechanism involved for this reaction and influence of reactor parameters on the reaction kinetics is unknown. This part of the research therefore aims to expand the insight on the mechanism involved for the electrochemical reduction of levulinic acid to valeric acid and furthermore, identification of critical reaction parameters to enable an optimal process design.

Figure 1. H-cell used for preparative electrolysis.

Using a novel three-compartment electrochemical reactor,[2] having a centre compartment filled with porous solid polymer electrolyte (PSPE), enables production of pure (salt-free) hydrogen peroxide. However, to come to an energy efficient electrochemical cell design, efficient integration of the electrocatalyst an optimal design of the porous solid polymer electrolyte is required. The aim of this part is to provide insight in 1. the critical parameters for efficient integration of electrocatalyst and 2. influence of the properties of the porous solid polymer electrolyte on the mass and energy transport in the electrochemical reactor.

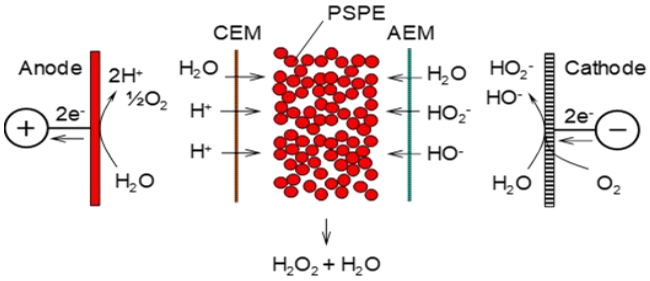


Figure 2. Cell design for the electrochemical production and in-situ concentration of H_2O_2.

Type of student projects envisioned

Experimentally oriented projects are available to improve understanding of the involved reactions, e.g. assessing the influence of various operational conditions and the synthesis and characterization of electrode materials. Opportunities for modeling projects are available in electrochemical process simulation.

[1] Bisselink et al., *ChemElectroChem* **2019**, 6, 3285.
[2] Bisselink (**2017**). Electrochemical process and reactor. (Patent No. WO2017222382)

Selective polysaccharide oxidation – new catalysts and new chains

Name PhD/PD: Konstantina-Roxani Chatzipanagiotou
Involved staff members: prof. dr. Harry Bitter
Project sponsor: Avebe
Start/(expected) end date of project: October 2019/August 2021

Background and goal of project

Starch is an interesting and widely available bio-based feedstock, which can replace fossil-based compounds in various applications, including the food industry, as well as the production of textiles, coatings, adhesives and biodegradable packaging materials. For further utilization, starch needs to be partially oxidized to anionic starch. Current oxidation methods in industry require harmful bleaching agents. Thus, to reduce the environmental impact of starch oxidation, new methods need to be explored. Here, a heterogeneous catalytic method is proposed, using a supported platinum-based catalyst, and oxygen gas as a sustainable oxidizing agent.

The objective of this project is to investigate the role of the catalyst's properties (support polarity, Pt particle size) and substrate properties (starch size, branching, and pre-treatment) for starch oxidation.

Highlight of the past year

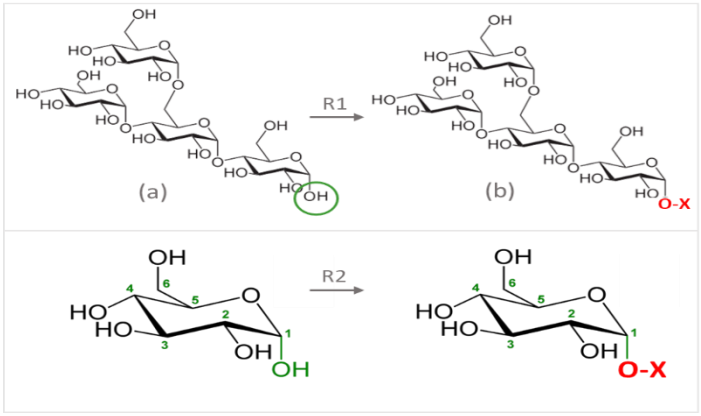


Figure 1: Selective pre-treatment (R1; i.e. reaction 1) of a starch molecule (a) that results in blocking of the terminal carbon (b). The same reaction (R2) has been previously shown for the glucose molecule.

We have previously shown that starch can be oxidized using oxygen over a Pt catalyst. However, the selectivity of this reaction is limited to the reducing terminal carbon of the polymer (highlighted with a green circle in Figure 1a). To achieve sufficient degree of oxidation for industrial application, the selectivity of this reaction needs to be stirred to other parts of the polymer.

Our preliminary results with glucose, the building block of the starch polymer, show that stirring the selectivity can be achieved by chemically blocking the terminal C1 carbon position (Figure 1, R2). Specifically, more oxidation was achieved on the C6 position of the molecule, when the C1 position was chemically blocked (for example by a methyl group). Following up on these results, we prepared a modified starch, for which the C1 position was blocked by a methyl group (Figure 1, R1). This blocking for starch polymers has not been shown before to the best of our knowledge.

Currently, more research is performed on optimizing the chemical pre-treatment reaction of starch and glucose. Furthermore, we are testing the effect of chemical pre-treatment on the selectivity of starch oxidation.

Type of student projects envisioned

This research project will be concluded in 2021. Therefore, no student projects are available within this topic.

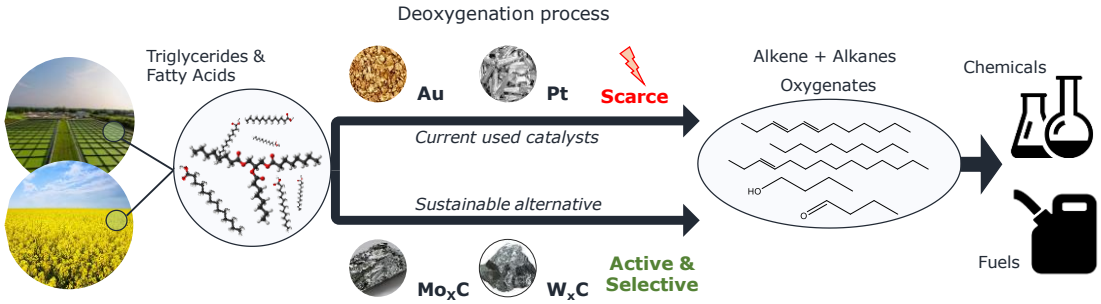
We would like to acknowledge the students involved in this project, namely Marlene Führer, Samuel Hijmans, and Luana Souza Macedo, for the valuable contribution to the experiments performed.

Mixed metal carbides for biomass upgrading

Name PhD/PD: Marlene Führer
Involved staff members: Prof. dr. Harry Bitter, dr.ir. Tomas van Haasterecht
Project sponsor: NWO & FAPESP
Start/(expected) end date of project: September 2018- September 2022

Background and goal of project

Transition metal carbides, such as those of molybdenum and tungsten, are considered viable alternatives to noble metal catalysts. It has been shown that these carbides were active for decarboxylation/ decarbonylation and hydrodeoxygenation of triglyceride-based feedstock to valuable alkenes and oxygenates. Interestingly, the supported W-carbides is more selective (>50%) towards the valued alkenes, while the supported Mo-carbide is more selective towards the oxygenates (30%). In our research we want to combine the performance properties of both catalyst by using mixed W/Mo-carbides.



Highlight of the past year

We showed this year that the Mo/W ratio in the bimetallic carbide-based catalyst influences the selectivity of the catalysts towards either oxygenates or alkenes. Figure 1 displays the conversion as function of oxygenates and alkene selectivity over mixed metal carbides. The mixed systems produced both, alkenes and oxygenates, in significant amounts. The mixed catalyst with higher Mo-content (3:1 Mo:W) has a slightly higher selectivity towards the oxygenates (25 mol%) and therefore resembles more the monometallic Mo-carbide. In comparison, the catalysts with higher W content (1:3) was more selective towards alkenes. Interestingly, the bimetallic catalysts retained high selective towards the alkene at higher stearic acid conversions (20-40 mol% alkenes at 90 % conversion), while the monometallic W-carbide shows lower alkene selectivity at higher conversions (10 mol% alkenes at 90% conversion). Thus the addition of molybdenum to W-carbide catalysts prevents full hydrogenation of the alkenes.

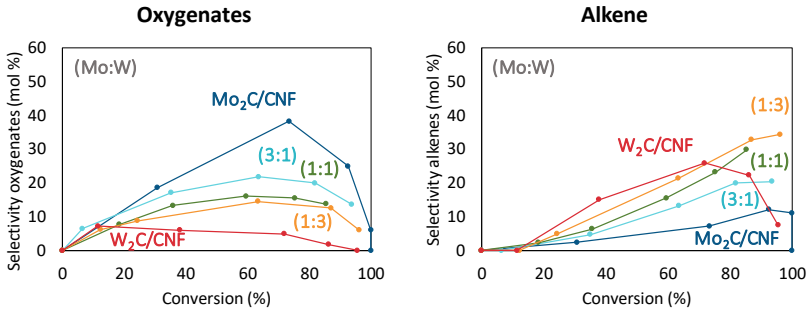


Figure 2. Conversion vs selective of oxygenates and alkenes of mixed metal carbides synthesized (250 mg catalyst, 2 g stearic acid, 50 mL solvent, 30 bar H_2, 350 °C).

Type of student projects envisioned

A thesis within this project involves mostly lab work including catalyst synthesis, characterization with techniques like XRD, TEM, N_2 physisorption and chemisorption. Sofar, the deoxygenation reactions are evaluated in a batch system. However, we are currently testing a plug flow system.

Processing of feathers to proteins – from fundamental insight to application

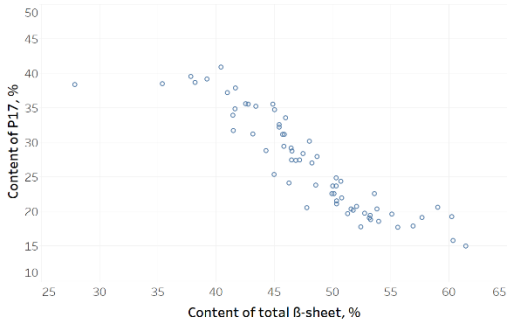
Name PhD: Xinhua Goerner-Hu
Involved staff members: Scott, E; Schneider, O; Haasterecht, T. van; Bitter,H
Project sponsor: SARIA International GmbH
Start(expected) end date of project: 2015/2021

Background and goal of project

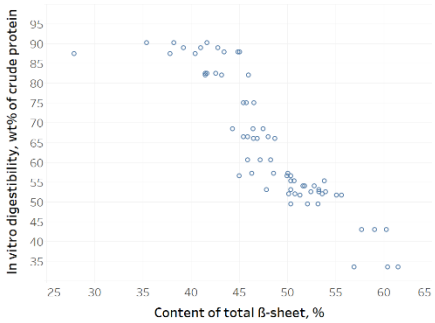
More than one million tons of feathers are produced as by-product annually in Europe. Feathers have a high protein content and contain various amino acids. Therefore, feathers have a good potential as feed ingredient. However, feathers are not digestible and have no nutritional value in their native state. Therefore, feathers have to be processed into more digestible proteins, peptides and amino acids. The processing changes the molecular structure of feathers, in particular, the secondary structure. The goal of our project is to relate the change of secondary structure to the processing conditions and to the effect of this change has on availability for enzymatic hydrolysis (AEH).

Highlight of the past year

Traditionally, AEH is studied by time consuming digestibility studies. We have shown that rather straightforward IR measurements of the processed feather samples gives a good prediction of that digestibility which makes sample analysis more facile. We observed a relationship between the □-sheet content and a new set of IR signals above 1700 cm-1 (left figure). In addition, we showed that the b-sheet content related to digestibility (right figure)

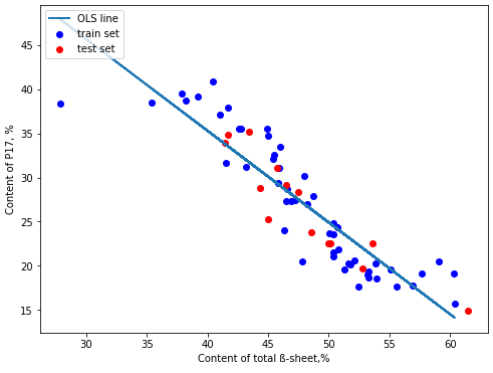


Relation □-sheet and new 1700cm-1 peak



Effect of ß-sheet change on in vitro digestibility

To validate the results between new peaks and b-sheet content, Python modeling (sklearn.linear_model, LinearRegression) was applied – 80% of data points were for model building (machine learning) and 20% of data points were for validation of built model. The model validation showed good result ($R2 = 0.88$, $p < 0.0001$).



Type of student projects envisioned

MSc student: lab work, e.g. experiment performance and analysis

Electro-conversion of biobased feeds into valuable platform chemicals

Name PhD: Matthijs van der Ham

Involved staff members: Prof. Dr. Harry Bitter (BCT-WUR)
Prof. Dr. Marc Koper (CASC-UL)

Project sponsor: NWO-TTW (perspectief), AVEBE, Brightlands and TNO

Start/(expected) end date of project: September 2019 - September 2023

Background and goal of project

Electrocatalytic conversions can aid in the sustainable conversion of biomass-based feedstocks, like glucose, starch and furans. Electrocatalysis uses electricity to drive conversions and water as oxidative or reductive agent. These features enables the use of renewable energy sources (e.g. solar or wind power) and circumvent the use of hazardous chemicals. Although electrocatalysis can aid in making the chemical industry sustainable, the research for electrocatalytic conversions remains limited to small molecules with few functional groups (like CO_2, N_2, H_2O) and bulk metal electrodes with low surface to volume ratios (Fig. 1A).

In my project I will focus on the electrocatalytic conversion of large molecules with many functional (e.g. hydroxyl, aldehyde and carboxylic acid) groups using supported catalysts (e.g. Pt/CNF) with high surface area too volume ratios. This will bring about new challenges, such as tuning 1) the reaction conditions to selectively convert the reactants to the desired products and 2) the design parameters of supported catalysts to avoid mass transport limitations (Fig. 1A+B). Therefore, the first project aims to research the effect of reaction conditions on the selective conversion of glucose to value-added products (e.g. gluconic acid), circumventing the formation of degradation and isomerization products like fructose and formic acid. The objective of the second part of the project is to build a model containing various design parameters, such as porosity and tortuosity for the electrocatalytic oxidation of glucose. This model be used to study the mass-transport phenomena of glucose in high surface area electrodes.

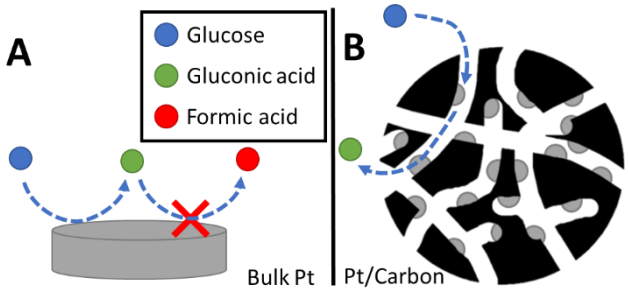


Figure 1. A) the system setup under which the effect of reaction conditions will be studied for the selective conversion of glucose and B) a representation for the model for high surface area electrodes (Pt/CNF), that will be used to study the effect of design parameters on the electrocatalytic oxidation of glucose

Highlight of the past year

Last year Pt on carbon nanofibers (Pt/CNF) catalysts were synthesized with different types and quantities of support oxygen groups on CNF. These catalysts were used to study the effect of support oxygen groups on the electrocatalytic oxidation of glucose. I found that these support oxygen groups can be tuned to increase the measured current (I) or reduce the applied potential (E) during glucose oxidation reactions, meaning that less energy input (W=I*E) is required to drive the reaction. Moreover, an increase in current is proportional to an increase in catalytic activity and therefore displays enhanced reaction rates. I also found a change in reaction mechanism, indicative for an effect on selectivity of the catalyst. The effect on catalytic selectivity is ongoing research.

Type of student projects envisioned

To steer the selective oxidation of glucose towards desired products the effect of various reaction condition (potential, pH, T, type of catalyst) needs to be studied. For this project I am looking for an enthusiastic student for labwork. To research how the design parameters affect the electrocatalytic oxidation of glucose in high surface area electrodes a model for mass-transport phenomena needs to be build. For this project I seek a motivated student that has experience with MATLAB or Python and is willing to build a model.

Selective polysaccharide oxidation – new catalysts and new chains

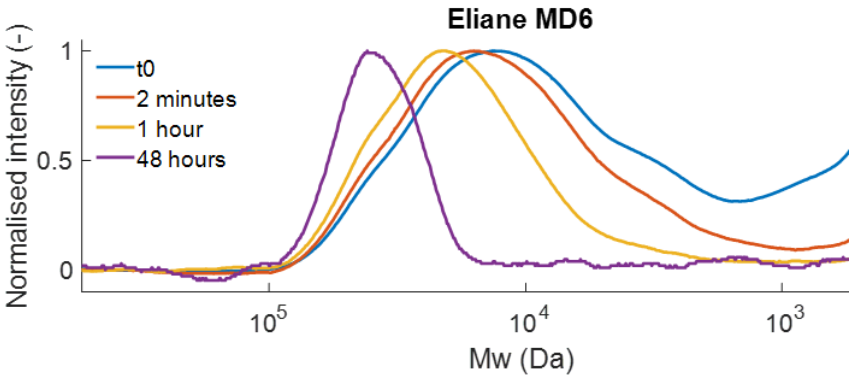
Name PhD: Tim Hoogstad
Involved staff members: Lars Kiewidt, Harry Bitter
Project sponsors: NWO, Avebe
Start/end date of project: 01-2016 till 12-2020 (Finished)

Background and goal of project

Oxidised starches are used as a biobased replacement for the petrochemically based polyacrylates and polyacrylamides. These oxidised starches have a broad range of applications. They are used as surface sizing agents in the paper and textile industry, as thickeners and texturisers in the food industry and have specialised applications in wastewater treatment, superabsorbents, and drilling fluids.
Oxidised starch is currently produced by mixing the starch with sodium hypochlorite (household bleach), however the large-scale use of hypochlorite has some undesired side-reactions (chlorate formation and depolymerisation), produces undesired by-products (salts), and has a high environmental impact.
In this project, the goal is to develop an alternative oxidation process that employs a heterogenous catalyst to facilitate the use of oxygen from air as oxidant. Hereby we aim to eliminate the use of hypochlorite from the process. My role in this project is to gain insight in how certain properties of the starch, specifically molecular size, influence diffusion, adsorption, and reaction rates in this conversion.

Highlight of the past year

One of the challenges in working with starch, is that it is polydisperse, meaning that there is always a mixture of differently sized starch molecules present in a reaction mixture. Through experiments, we have gained insight in how these distributions of starch molecules of different sizes adsorb on the catalyst support material. We've observed that the adsorption process is size-selective, and that smaller molecules (<5kDa) adsorb at an approximately 50% higher rate than the large molecules (>50kDa). As a result, the number average molecular weight of starch in solution increases approximately a factor 3 over 48 hours, thus showing a dynamic substrate distribution. This type of knowledge allows us to predict the size distribution of the formed product and aids in the further development and optimization of a catalyst for the air oxidation of starch.



Type of student projects envisioned

Considering the end-date of the project, no student projects are available under my supervision. However, closely related research is still ongoing in the group. If you're interested in this topic, please contact Roxani Chatzipanagiotou or Matthijs van der Ham for further information.

Selective catalytic transformations of non-edible carbohydrates

Name PhD: Frits van der Klis
Involved staff members: Prof. Dr. J.H. Bitter; Dr. D. S. van Es; Dr. J. van Haveren
Project sponsor: TKI-programs, CatchBio, EU SPLASH, EU Pulp2Value
Start/(expected) end date of project: October 2013 – October 2021

Background and goal of project

The aim of my research is to convert non-edible carbohydrates found in waste food crop residues, like sugar beet pulp, into useful chemicals and (polymer) materials. Carbohydrate structure plays a pivotal role during catalytic conversions of these feedstocks, and forms the core of this research.

Highlight of the past year

This year was dedicated to finalize my thesis, which deals with the following topics:
[1]: The first part of my thesis focusses on the conversion of non-edible C5-sugars (xylose and arabinose) into C4-tetritols (low-calorie sweeteners and chemical building blocks) over ruthenium catalysts. A high selectivity towards C4-tertritols was achieved under mild aqueous conditions, and it was found that the mechanism for C5-sugar conversion involves a formal decarbonylation step of which the reaction rate depends on the carbohydrate structure.[1]
[2]: The second part focusses on the gold-catalysed oxidation of galacturonic acid to galactaric acid. This product can be used for e.g. corrosium inhibition or again as a chemical building block. Gold catalysts showed excellent selectivity and stability, and the use of a continuous flow reactor was found more productive compared to a batch process.[2] We are currently investigating the influence of the carbohydrate structure on the oxidation rate, as well as the influence of reaction conditions on the reaction mechanism.
[3]: In the third part, a novel strategy was developed to convert galacturonic acid, via a catalytic isomerization step, into furan-2,5-dicarboxylic acid (FDCA). FDCA is a biobased alternative for the petrochemical-based terephthalic acid, the main building block for PET-bottles.[3]
[4]: In the final part, building blocks like FDCA were combined with sugar-based 1,4-butanediol derivatives. This work focused on the influence of methyl-substituents on 1,4-butanediol-analogues on the final polymer properties. The number of methyl groups was found to increase the glass transition temperature (Tg) with increasing numbers of methyl-substituents. The chirality of these methyl-groups influences the crystallization behavior of the polymers, and opens possibilities for the development of new stereocomplex materials, as well as possibilities for tuning of various (co)-polyester properties.

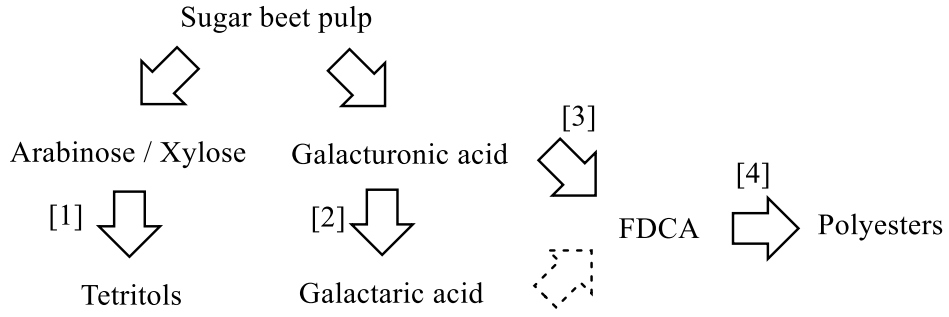


Fig. 1: Overview of research topics; [numbers] correspond to text- and reference numbers.

Type of student projects envisioned

Student projects all involve organic chemistry and/or catalysis orientated lab work, focused on the conversion of carbohydrates. Standard analysis during synthesis includes NMR, GC-MS, IR and HPLC. Catalysts will be analyzed by TEM, XRD, chemisorption and physisorption.

[1] *Green Chem.*, **2015**, *17,* 3900 – 3909; [2] *React. Chem. Eng.* **2018**, *3*, 540 – 549; [3] *ChemSusChem*, **2017**, *10*, 1460-1468; [4] *J. Polym. Sci., Part A: Polym. Chem.,* **2018**, *56*, 1903-1906.

Specific dietary fiber combinations for decreasing antibiotics use and faster recovery of gut microbiota

Name PhD: Cynthia Klostermann, MSc
Involved staff members: prof. dr. Harry Bitter
Involved members: prof. dr. Henk Schols (FCH), prof. dr. Paul de Vos (UMCG)
Project sponsor: NWO, CCC (CarboBiotics)
Start/(expected) end date of project: 15-11-2018 / 15-11-2022

Background and goal of project

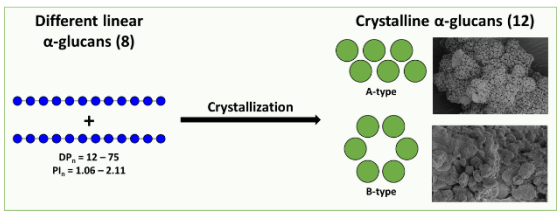
Antibiotics have a negative effect on beneficial microbiota and gut barrier function. Because of these negative effects and the fact that bacteria become more and more resistant against antibiotics, there is a need for reduction of antibiotics use. One possible alternative for antibiotics might be special developed fibers that stimulate beneficial microbiota and positively stimulate immune barrier function. Resistant starch type 3 (RS-3) might be such a fiber, since it is fermented by microbes that produce butyrate. Butyrate is known to be very important for gut health.

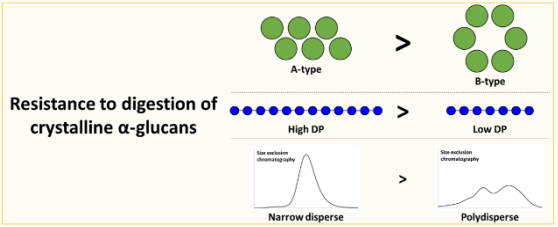
The main questions we try to answer in this project are:

1) Which characteristics of RS-3 makes it resistant to digestion in the small intestine?
2) If RS-3 arrives in the colon, which microbes degrade it and how? What fermentation products are formed?
3) Do gut microbiota that were treated with antibiotics recover faster in the presence of RS-3?

Highlight of the past year

Twelve unique RS-3 samples were produced, which differed in average Mw, Mw distribution and crystal type. The twelve RS-3 samples were in vitro digested and results showed that crystal type is the most important factor to reduce digestibility; A-type crystals are much more resistant to digestion than B-type crystals. Also Mw and Mw distribution effects on digestibility were shown. Next, the RS-3 crystals were in vitro fermented using batch fermentation and fecal inoculum of piglets and infants. Also here, results have shown big differences between degradation and fermentation of A- and B-type crystals by fecal microbiota.





Student projects envisioned

Research topics involve lab work on chemical and physical characterization of the different dietary fibers (GOS, IMMPs, RS-3) (eg. HPAEC-PAD, HPSEC-RI, 1H-NMR, XRD, DSC, Mastersizer). In addition, fermentation studies using these fibers and fecal inocula of piglets and infant/adults might be studied.

Development of bifunctional catalysts for CO2 conversion to chemicals

Name PhD/PD: Nazila Masoud
Involved staff members: Harry Bitter
Project sponsor: -
Start/(expected) end date of project: October 2019-October 2021

Background and goal of project

Excess carbon dioxide (CO_2) in the atmosphere causes environmental concerns. To mitigate negative effects, CO_2 needs to be captured from air, and stored safely or utilized sustainably. Hence, an abundant carbon source to produce chemicals in a circular economy becomes available after CO_2 capture from air. CO_2 can be utilized to produce chemicals. However, CO_2 is thermodynamically very stable. Hence, it must be reacted with high energy molecules like hydrogen, alcohols, olefins or epoxides. This is an activated process that needs a catalyst.

The CO_2 that is captured from air often coexists with O_2, since full separation of CO_2 from O_2 requires high energy input. Hence, it is necessary to develop reactions that can occur in oxidizing atmospheres and catalysts that are efficient and stable under these conditions. This project aims to design bifunctional catalysts to convert CO_2 to organic molecules under oxidizing atmospheres (Figure 1). One function of the catalyst needs to activate the CO_2 with minimum energy input. The other function needs to bind the reactive molecule in proximity of the CO_2, to allow the reaction to occur. We apply solid bases e.g. magnesium oxide that activate acidic CO_2 and mix them with solid acids e.g. Al_2O_3 that interact with a second molecule like an epoxide. These oxides are highly stable under oxidizing atmospheres. The first step is to find basic oxides that have interaction with the CO_2 in an optimum strength. The second step is to find an optimum composition of the mixed oxides, with respect to basic/acid strength and spatial distance, for a desired reaction to occur.

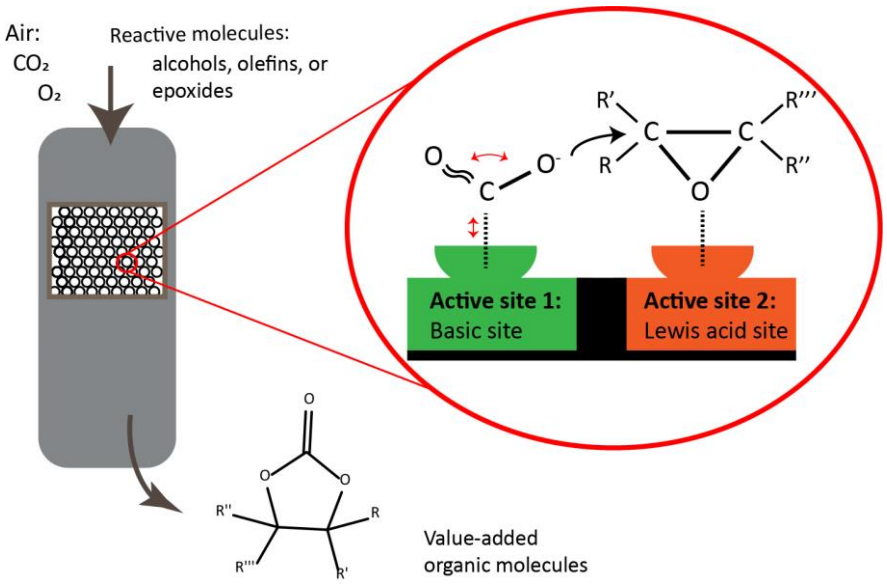


Figure 1. Rational design of bifunctional solid catalysts to convert CO_2 to organic molecules

Highlight of the past year: We applied Infrared spectroscopy to investigate interaction of different solid bases with CO_2. We examined solid bases such as CaO, MgO, and ZnO as a catalyst to activate CO_2 in reaction with styrene oxide to produce styrene carbonate. We extended this examination to hydrotalcites which are mixtures of MgO and Al_2O_3 with different Mg/Al ratios. We learned that the presence of both basic and acid sites in an optimum composition is necessary for higher yield and selectivity of the reaction. Side reactions in the presence of O_2 is still an issue to be resolved.

Type of student projects envisioned: The project includes chemical synthesis of inorganic nano materials and application of different chemisorption and spectroscopy techniques.

Characterization of surface events by calorimetry

Name PhD/PD: Nazila Masoud
Involved staff members: Tomas van Haasterecht and Harry Bitter
Project sponsor: -
Start/(expected) end date of project: October 2019-October 2021

Background and goal of project

Calorimetry is the measurement of heat transfer associated with chemical reactions, physical changes, or phase transitions under specific conditions. It is a technique to study catalysts, supports, sorbents, and for the characterization of their surfaces *in situ* via following the associated heat release/consumption. We aim to develop applications of calorimetric techniques to investigate surface events during catalytic reactions. We apply a C80 microcalorimeter that is equipped with different accessories to study gas phase as well as liquid phase catalytic reactions e.g. biomass conversions.

Our equipment, i.e. a C80 uses a Calvet calorimetric detector in which a sample and reference cell are totally surrounded by an array of thermocouple detectors allowing for the measurement of all heat released or consumed in high accuracy (Figure 1). The sample cell contains the catalyst/sorbent powder under study. Both cells are exposed to the same reactive media in gas/liquid phase and the associated heat release or consumption is indicator of the interaction of the sample with the reactive media.

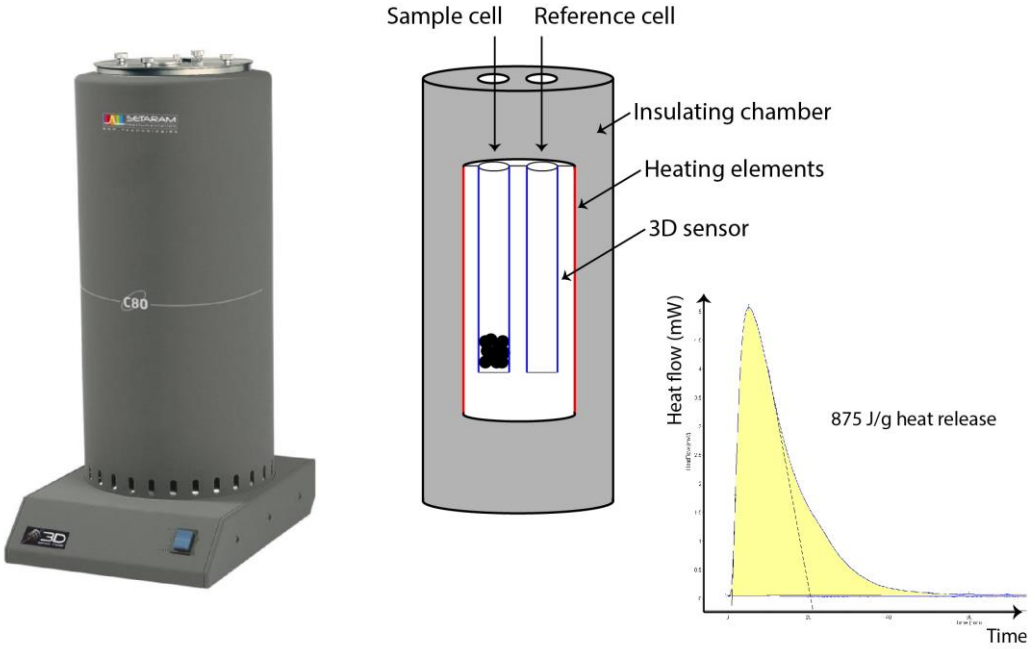


Figure 1. A photo of C80 calorimeter (left), illustration of its components (middle), and a graph showing heat release during CO_2 capture from air upon exposure to supported K_2CO_3 based sorbent (right)

Highlight of the past year

The C80 calorimeter was commissioned. Experiments were performed to measure heat release during CO_2 capture from air over carbon supported K_2CO_3 sorbent (Figure 1). This study showed that the main contribution of heat release/consumption is due to wetting the sorbent or water evaporation rather than carbonation and decarbonation of the sorbent.

Type of student projects envisioned

This project includes lab work and requires knowledge in physical chemistry.

Catalyst development by physiochemical modification of graphite

Name PhD/PD: Nazila Masoud
Involved staff members: Tomas van Haasterecht and Harry Bitter
Project sponsor: NWO-KIEM
Start/(expected) end date of project: October 2019-October 2021

Background and goal of project

Graphite is an allotrope of carbon. It is a crystalline material consisting of multiple graphitic layers. In each layer, carbon atoms in sp^2 hybridization are arranged in a honeycomb lattice. Layers are bonded to each other via weak van der Waals forces. These week forces allow layers of graphite to be easily separated, or to slide past each other by shear stress, so called exfoliation. The graphite structure can also be broken by mechanical forces, so called fragmentation. Exfoliation and fragmentation increase graphite's surface area. Fragmentation also creates smaller fragments of graphitic structure with reactive sites on the edges. By adding different chemicals in different concentrations during fragmentation/exfoliation, these active sites can be functionalized. In this way Edge-modified-nano-graphite (EMNG) can be produced (Figure 1).

EMNG has potential applications in catalysis because of their high surface area and (introduced) functionalities. This project aims to prepare EMNGs from waste graphite using a rotary ball mill. A rotary ball mill applies shear stress and mechanical forces. Chemical additives are being used to modify the graphitic structure and to put selective surface functions on the graphite. Prepared samples are characterized by different chemisorption and spectroscopy techniques and are examined for selective catalytic reactions.

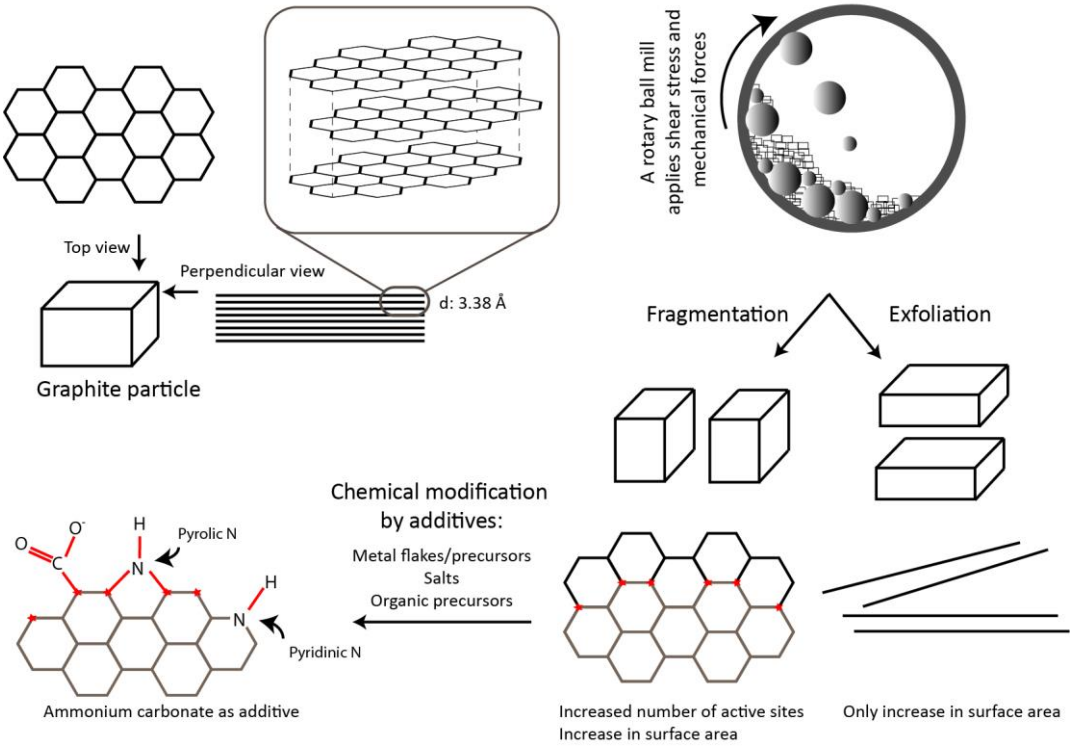


Figure 1. Illustration of the graphite structure and its physiochemical modification by a rotary ball mill

Highlight of the past year: We successfully prepared graphite with surface area as high as 500 m^2/g, however controlled addition of functional groups was a challenge.

Type of student projects envisioned: The project includes working with a pilot rotary ball mill, characterization of the EMNGs, and evaluation of their performance in different catalytic reactions.

(In situ) charachterization of tungsten and molybdenum carbides

Name PhD/PD: Edwin Schreuder
Involved staff members: Prof. dr. Moniek Tromp (RUG), Prof. dr. Harry Bitter
Project sponsor: RUG/WUR
Start/(expected) end date of project: February 2020 – February 2024

Background and goal of project

Transition metal carbides, especially tungsten and molybdenum carbides are known to be good candidates to replace the traditionally used noble metal, which are scarce, for a variety of chemical reactions. Tungsten carbide and molybdenum carbide can exist in a variety of phases ranging from higher carbides (MoC, WC) to lower carbides (Mo_2C, W_2C) with structures in between (WC_{1-x}, MoC_{1-x}). Furthermore, these structures are all prone to oxidation in reaction environments, further increasing the amount of phases to be taken into account during catalysis.

Since initial research suggests that the catalytic behaviour of these carbides is influenced by which phase is present,[1] thorough characterization is needed. The goal of this project is to investigate the detailed structure and electronics of the different carbides and investigate what happens with the catalyst in reaction environment (*in situ*) and during a reaction (operando) by using X-ray techniques like XRD and X-ray absorption and emission spectroscopy (XAS and XES).

Highlight of the past year

The first tungsten carbide samples were synthesized on carbon nanofiber support and characterized by X-ray Absorption Near Edge Spectroscopy (XANES). XANES manages to distinguish two different compounds among the three different samples. This was confirmed by XRD, which identified these two phases as W_2C and WO_3. As the differences in the XANES spectrum are small, further XANES, but also XES measurement are needed to determine the limitations of the techniques.

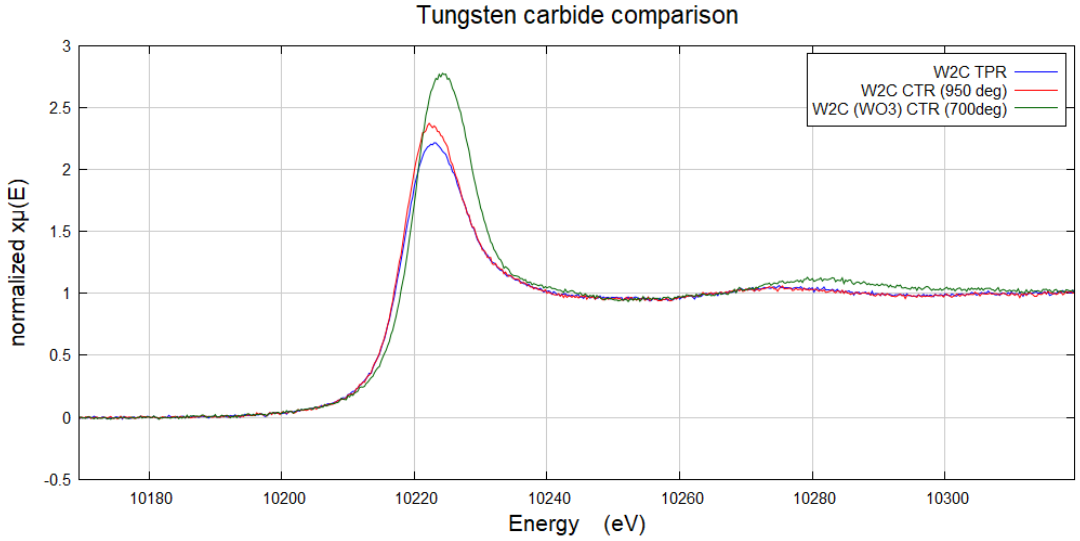


Figure 1: XANES measurement of three different samples: W_2C synthesized by carbothermal reduction (CTR) at 950°C (red), WO_3 created by incomplete CTR at 700°C (green); W_2C prepared by thermal programmed reduction (TPR) at 700°C under 80/20 H_2/CH_4 flow (blue).

Type of student projects envisioned

Student projects could involve labwork mainly consisting of synthesis and characterization of a wider variety of carbides (doped, different support) or different tungsten compounds (nitrides, sulfides).

1. Stellwagen, D. R. & Bitter, J. H. Structure-performance relations of molybdenumand tungsten carbide catalysts for deoxygenation. *Green Chem.* **17**, 582–593 (2015).

Bioelectrochemical chain elongation of CO2 to caproate:
Electrification of biotechnology

Name PhD: ir. Sanne de Smit
Involved staff members: prof. dr. Harry Bitter (BCT), prof. dr. ir. Cees Buisman (ETE), dr. ir. David Strik (ETE)
Project sponsor: WIMEK, ChainCraft
Start/(expected) end date of project: Sep 2018 – Sep 2022

Background and goal of project

A very important climate goal is the reduction of CO_2 emissions. Carbon capture and utilisation projects facilitate this goal. Carbon dioxide can be elongated by microorganisms in a so-called chain elongation process. Some microorganisms can use sole electrons as energy input for this process, they grow as a biofilm on a cathode in a bioelectrochemical system (Figure 1). Products of the conversions are fatty acids, which can be used as animal feed additive or platform chemical for e.g. fuel production.

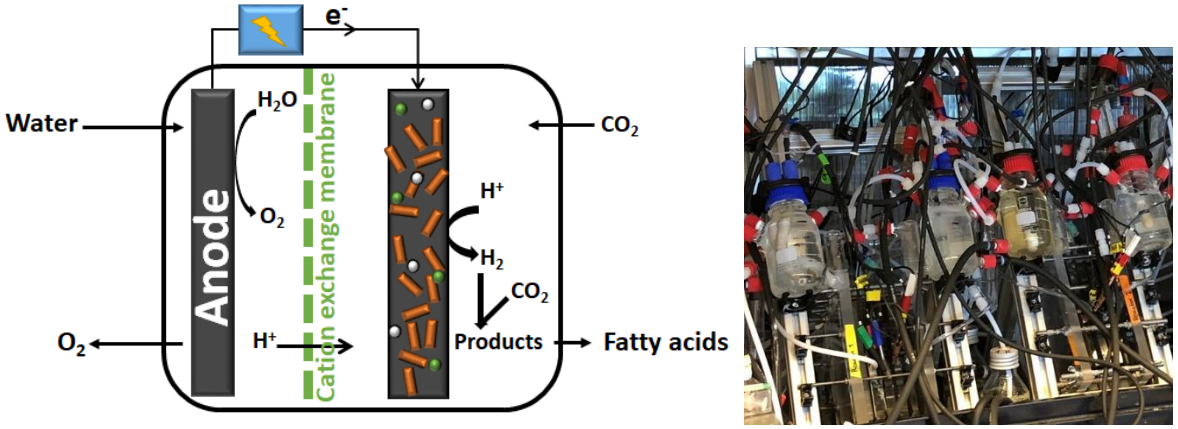


Figure 3. *Schematic (left) and actual (right) setup of bioelectrochemical chain elongation systems.*

The goal of the PhD project is to study the role of the biofilm and the changes therein in the optimization of the electron flux between the cathode and the biofilm. Gaining insights on the working mechanisms will provide steering tools to optimize the system in the lab and work towards practical application.

Highlight of the past year

We found that the reactor current can be increased by performing the analysis technique "Cyclic Voltammetry" (CV). Together with the current increase, metal concentrations increased in the reactor liquid. This indicates that the release of these metals from the cathode correlates to the current increase. The current increase would be beneficial for the system, since it causes a higher energy availability for the bacteria. To further develop this CV as tool to boost the performance, the metal behaviour on the cathode is studied.

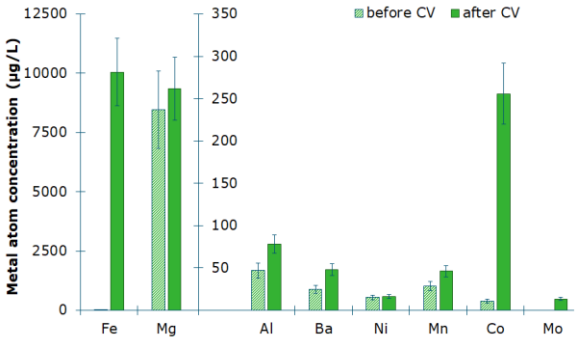


Figure 4. *Metal concentrations in the reactor liquid before and after cyclic voltammetry (CV).*

Type of student projects envisioned

A student on this project would work in the laboratory, maintaining and analysing the bioelectrochemical systems. The experimental conditions can be varied to study different parts of the microbial and electrochemical processes occurring at the cathode. Analysis of the system is done using chromatography, microscopy, sequencing of the microbial community and spectometry. Model studies can be performed to understand underlying chemical or thermodynamic mechanisms.

Updating the online Micromasters: Chemistry and Technology for Sustainability

Name PD: Dr. Carlos Cabrera
Involved staff members: Prof. dr. Harry Bitter
Start/(expected) end date of project: Nov 2019 - Nov 2021

Background and goal of project

The micromasters "Chemistry and Technology for Sustainability" is an online course part of WUR-Edx platform. This Micromaster consists of three interrelated courses, taking the students all the way from how to switch from fossil resources to biomass, to build an efficient and sustainable biorefinery and finally they dive to design an advanced biobased conversion. Each course take the student through all the important aspects required to build a Biobased Economy. The courses are:

a) Fossil resources to biomass- a business and economic perspective
b) Biorefinery- from biomass to building blocks of biobased products
c) Advanced biobased conversions

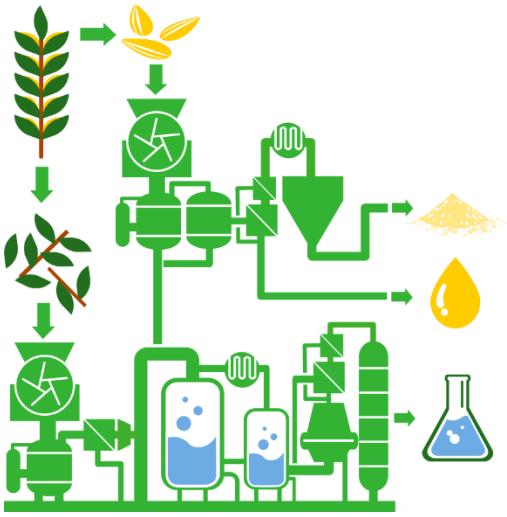


Figure 1. Biorefinery scheme taken from course: Biorefinery: from biomass to building blocks.

Highlight of the past year

During the last year, an exam bank was created and implemented to have multiple exam questions for two of the courses of the micromaster (Chem01 and Chem02). Additionally, several questions were improved to match the learning objectives of the courses.

Physical Chemistry - Biobased Soft Materials

Team Leader: Dr. Costas Nikiforidis

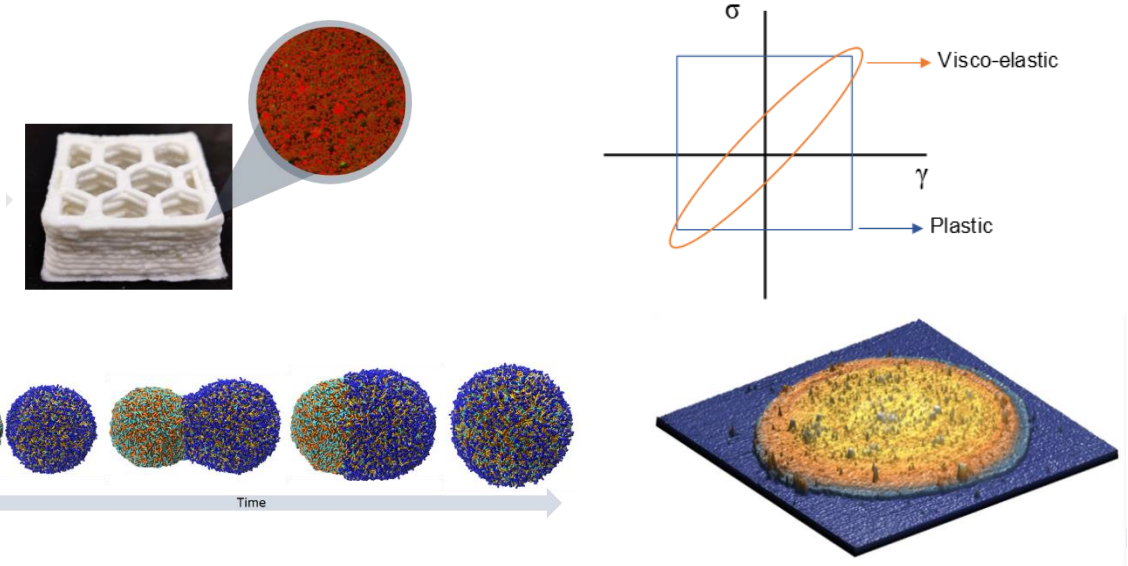
PhD students: Eleni Ntone, Simha Sridharan, Laura Schijven, Umay Sevgi Vardar-Kule, Mingzhao Han, Zhaoxiang Ma, Lorenz Plankensteiner, Kübra Ayan

Contact: costas.nikiforidis@wur.nl

Background and goal

The Biobased Soft Materials team is interested in understanding the interactions in natural supramolecular architectures to construct new bioderived and bioinspired soft materials for practical uses. The self-assembly and equilibrium properties of the soft materials are investigated from a soft matter physics, chemistry, and biology perspective at multiple length scales, from molecular to macroscopic.

The typical analytical tools we are using in our research are atomic force, confocal, scanning and transmission electron microscopy, static and dynamic light scattering, interfacial, and bulk rheology.



Main topics

- Protein cages to carry therapeutics and diagnostics
- Trafficking molecules with Lipid Droplets (Oleosomes)
- Behavior of Lipid Droplets (oleosomes) on interfaces
- Plant protein mixtures as emulsifiers
- Jammed emulsions for 3-D printing
- Design bio-inspired oil droplets resistant to lipid oxidation
- Extraction of proteins and Lipid Droplets using their mobility under an electric field

Highlights from last year

-ACS Author Choice in Langmuir (August - October 2020) - 10.1021/acs.langmuir.0c01955
-Most Downloaded research article in Food Hydrocolloids (April - October 2020) - 10.1016/j.foodhyd.2019.105533

Students project envisioned

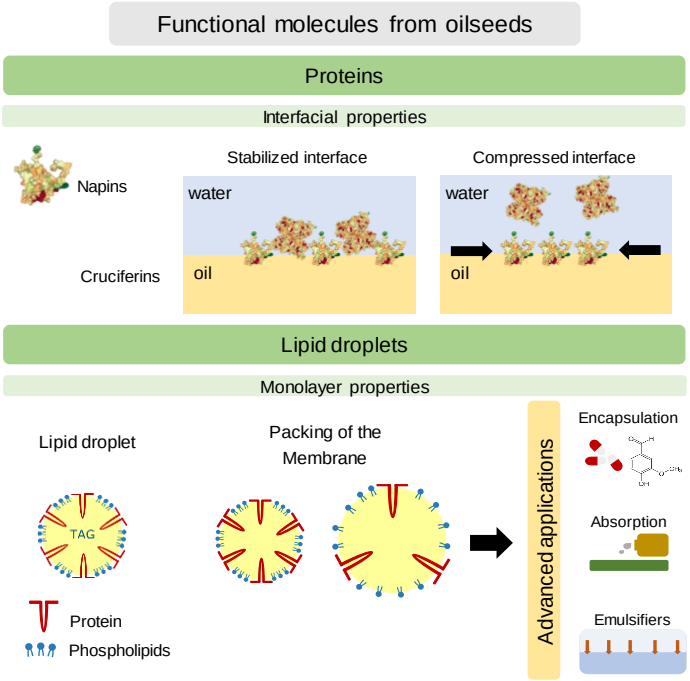
Thesis subjects are related to the research of the Ph.D. projects of the team including mainly experimental work and in some cases Molecular Dynamic Simulations

Plant based soft materials for advanced applications

Name PhD: Eleni Ntone
Involved staff members: BCT: Costas Nikiforidis, Harry Bitter, **FPH:** Leonard Sagis
Project sponsor: NWO-TIFN
Start/(expected) end date of project: September 2017/September 2021

Background and goal of the project

In this project we use experimental soft matter science to investigate the interfacial properties of plant derived molecules, with focus on oilseed proteins and lipid droplets (oleosomes).
The first aim of the project was to obtain the proteins from the seed matrix with a simple treatment route. This process resulted in supramolecular protein extracts. Thereafter, we investigate the interactions between proteins and proteins and non-protein molecules in these complex systems at model interfaces. The upper aim is to determine the ability of these complex systems to be used in soft materials like emulsions and filled gels.
Oilseeds also contain a plethora of lipid droplets. Lipid droplets are cell organelles present not only in oilseeds but in cells from humans to insects. Their role is to traffic and channel lipids in the cells and they owe this ability to their unique phospholipid monolayer structure. Here, we purify lipid droplets and study the rheological properties of their unique monolayer and the ability of lipid droplets to absorb and release lipids on demand. This knowledge could be used to tune the monolayer properties of lipid droplets and exploit them in multiple advanced applications, like absorption and carrying of sensitive molecules.



Highlights of the past year

Rapeseed proteins present in mildly purified mixtures can stabilize interfaces and can be used in soft materials like emulsions and emulsion filled gels. The stabilization mechanism is pH dependent. Lipid droplets have an elastic dilatable monolayer that allows them to adsorb and release lipids, realizing lipid trafficking

Type of student projects envisioned

No available projects at the moment due to finalizing of the project

Related publications

- Ntone, E., Bitter, J. H., & Nikiforidis, C. V. (2020). Not sequentially but simultaneously: Facile extraction of proteins and oleosomes from oilseeds. Food Hydrocolloids, 102, 105598.
- Ntone, E., van Wesel, T., Sagis, L., Meinders, M., Bitter, J., & Nikiforidis, C. (2020). Adsorption of rapeseed proteins at oil/water interfaces. Janus-like napins dominate the interface. Journal Of Colloid And Interface Science, 583, 459-469.

Pea proteins as structuring agent in edible soft materials

Name PhD/PD: Lakshminarasimhan (Simha) Sridharan
Involved staff members: Dr. Costas Nikiforidis, Prof. Harry Bitter, Dr. Marcel B.J. Meinders
Project sponsor: TiFN, Wageningen
Start/(expected) end date of project: 02/10/2017-02/10/2021

Background and goal of project

Due to concerns over the negative societal and environmental impact of animal sourced foods, plant-based foods are gaining interest. Specifically, plant proteins are versatile biopolymers that can be used to create structures in edible soft materials. Plant proteins such as pea proteins can function as emulsifiers and gelling agents. Their functionality depends on the environmental conditions such as pH. To utilize pea proteins as functional agents, their functionality in relation to their physico-chemical properties need to be well understood. Therefore, this project focusses on understanding the emulsifying properties and general use in soft matter of pea proteins.

Highlight of the past year

- Pea proteins at acidic pH (pH 3) form a mixture of protein particles and protein molecules
- From the mixture of protein particles and molecules, the protein molecules stabilize oil droplets, while protein particles are present in the bulk
- Pea proteins are able to stabilize high oil emulsions (jammed emulsions) without the aid of any additional emulsifiers.
- At pH 3, pea protein particles cross-link the jammed oil droplets to form elasto-plastic 3D printable material

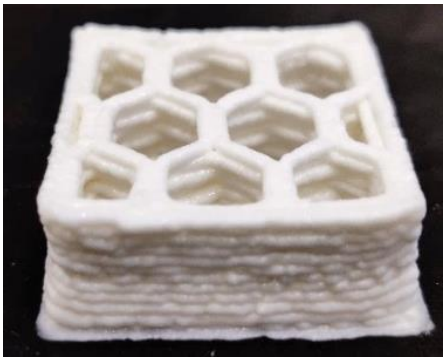


Figure 5: 3D printed honeycomb structure of pea protein stabilized jammed emulsion

Type of student projects envisioned

Currently no student projects are envisioned. Finishing date in Autumn 2021

Published articles:

- Sridharan, S., Meinders, M.B., Bitter, J.H. and Nikiforidis, C.V., 2020. Pea flour as stabilizer of oil-in-water emulsions: Protein purification unnecessary. *Food Hydrocolloids*, *101*, p.105533.
- Sridharan, S., Meinders, M.B., Bitter, J.H. and Nikiforidis, C.V., 2020. On the emulsifying properties of self-assembled pea protein particles. *Langmuir*, *36*(41), pp.12221-12229.

Gelation of egg yolk HDL by Au3+ ions

Name PhD: Laura Schijven
Involved staff members: dr. Costas Nikiforidis (BCT), Prof. dr. Harry Bitter (BCT), dr. Vittorio Saggiomo (BNT), Prof. dr. Aldrik Velders (BNT)
Project sponsor: VLAG Graduate School
Start/(expected) end date of project: September 2017 – June 2022

Background and goal of project

In conventional protein gelation, proteins (partially) unfold through heating. During protein unfolding, interior amino acids get exposed and cross-link with neighboring proteins by physical interactions and/or chemical bonds. Subsequently, those proteins can form a three-dimensional gel network.

Metal ions can also induce protein-protein interactions, such as aggregation and denaturation. In this project, we study the potential of using metal ions, in particularly Au^{3+}, to induce the formation of a protein gel network. Egg yolk high-density lipoprotein (HDL), a natural assembly of proteins and lipids, is used as a model protein.

Highlight of the past year

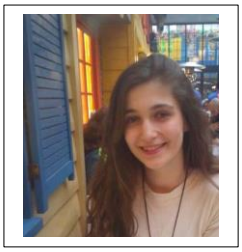
- HDL interacts with Au^{3+} ions at room temperature and it instantly forms a gel network.
- Additionally, Au^{3+} ions get reduced to Au^{0} and form AuNPs.
- Properties of the Au-HDL gels can be tuned by varying conditions, such as concentration, pH, temperature and ionic strength.

Type of student projects envisioned

Available student projects include lab work, focusing on physical and chemical characterization of interactions between Au^{3+} ions and HDL. The Au-HDL gels can find potential applications in materials and biological sciences, such as carriers of therapeutics or imaging agents. Main techniques include UV-Vis, DLS, NMR, AFM, CLSM and TEM.

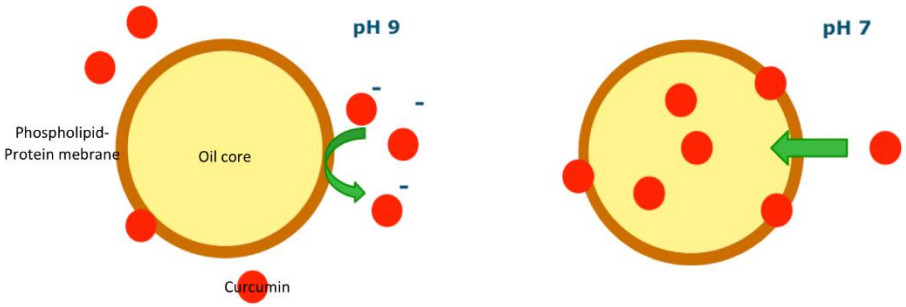
Loading of Lipid Droplets (Oleosomes) with curcumin

Name PhD/PD: Umay Sevgi Vardar-Kule
Involved staff members: Costas Nikiforidis, Harry Bitter
Project sponsor: Republic of Turkey Ministry of Education
Start/(expected) end date of project: 09.2019-09.2023



Background and goal of project

Lipid Droplets (Oleosomes) are natural spherical organelles, which are comprised by a hydrophobic triacylglycerol core, surrounded by a monolayer of phospholipids and proteins. They are present in all eukaryotic organisms and their biological role is to protect and channel lipids[1].
Additionally Lipid Droplets serve as carriers of functional biological compounds, like cholesterol, fat soluble vitamins etc., which are provided to the organism when needed[2]. In the light of this knowledge, the main purpose of the project is to investigate the natural function of oleosomes to carry hydrophobic molecules with the purpose of developing a green and natural carrier that could be used an alternative to synthetic lipid-based carriers in food or medical applications.



Loading mechanism of curcumin into Lipid Droplets

Highlight of the past year

- Curcumin was loaded into Lipid Droplets
- The diffusion of curcumin into Lipid Droplets driven by hydrophobic forces

Type of student projects envisioned

Available projects for students are included lab work :

- Tracking curcumin when diffused into Lipid Droplets,
- Physicochemical stability of Lipid Droplet membrane

For these projects, dynamic light scattering, UV-Vis. SDS-PAGE and Confocal microscopy will be extensively used.

1. Nikiforidis, C., Matsakidou, A. & Kiosseoglou, V., 2014. Composition, properties and potential food applications of natural emulsions and cream materials based on oil bodies. The Royal Society of Chemistry , p. 25067–25078.
2. Nikiforidis, C., 2019. Structure and functions of oleosomes (oil bodies), Advances in Colloid and Interface Science

Exploring the sustainable transformation of the plant extract industry

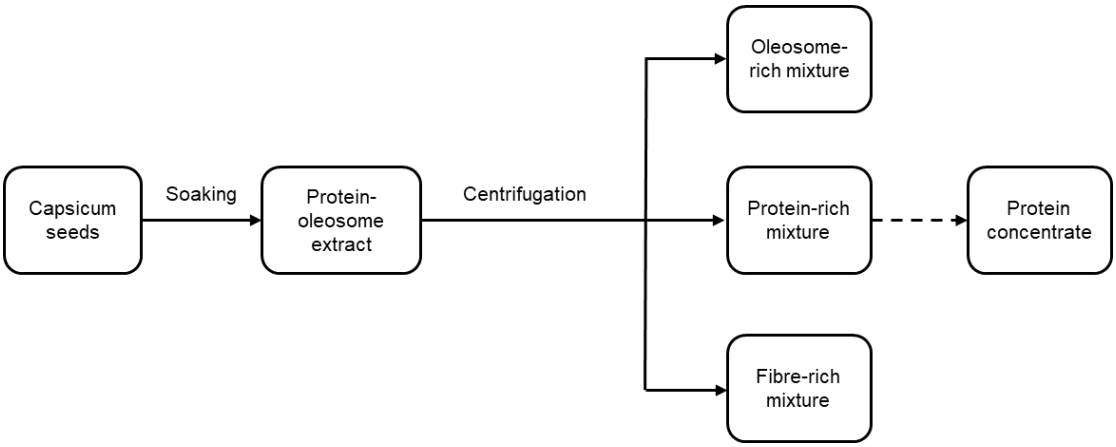
Name PhD/PD: Mingzhao Han
Involved staff members: dr. Costas Nikiforidis (BCT), Prof. dr. Remko Boom (FPE), dr. Xueqin Zhu (ENR)
Project sponsor: China Scholarship Council (CSC)
Start/(expected) end date of project: September 2019-September 2023

Background and goal of the project

The growth of the global human population to 9-10 billion people will put a strain on our possibilities to produce sufficient food, without depleting our natural resources in 2050s. In agriculture, modern genetic techniques and scientific framing have been applied to produce sufficient food for the current 7.5 billion people (Nikos Alexandratos and JelleBruinsma 2012). However, the conversion of agricultural raw materials into ingredients in edible products is generally not very efficient and potentially valuable feedstocks are being wasted(McClements 2020).

In our work, we are trying to valorise the side stream from the production of pigments from capsicum peppers. Chenguang Biotech Group Co., Ltd (CCGB). CCGB is the largest capsicum pigments producer in the world and produces significant side streams that are currently used as animal feed or burned. Capsicum seeds contain around 23 wt% oil and 21 wt% protein. We are aiming to aqueously extract the proteins and oil in the form of Lipid Droplets (oleosomes) and compare the costs of this process with the conventional way, where the oil and protein are extracted by applying an organic solvent extraction.

Finally, we will quantify the economic relationship between the inputs and the outputs in the conversion of the capsicum seeds into edible and valuable products.



Highlight of the past year

This project is started in WUR in September 2020 and is in collaboration with the Chinese Agricultural University (CAU)

Type of student projects envisioned

Student projects include lab work (extraction and characterization of oleosome-rich and protein-rich mixtures from capsicum seeds), and scaling up the oleosomes production process.

McClements, David Julian. 2020. "Development of Next-Generation Nutritionally Fortified Plant-Based Milk Substitutes: Structural Design Principles." *Foods* 9(4):421.

Nikos Alexandratos and Jelle Bruinsma. 2012. *World Agriculture towards 2030/2050: The 2012 Revision Global Perspective Studies Team FAO Agricultural Development Economics Division*.

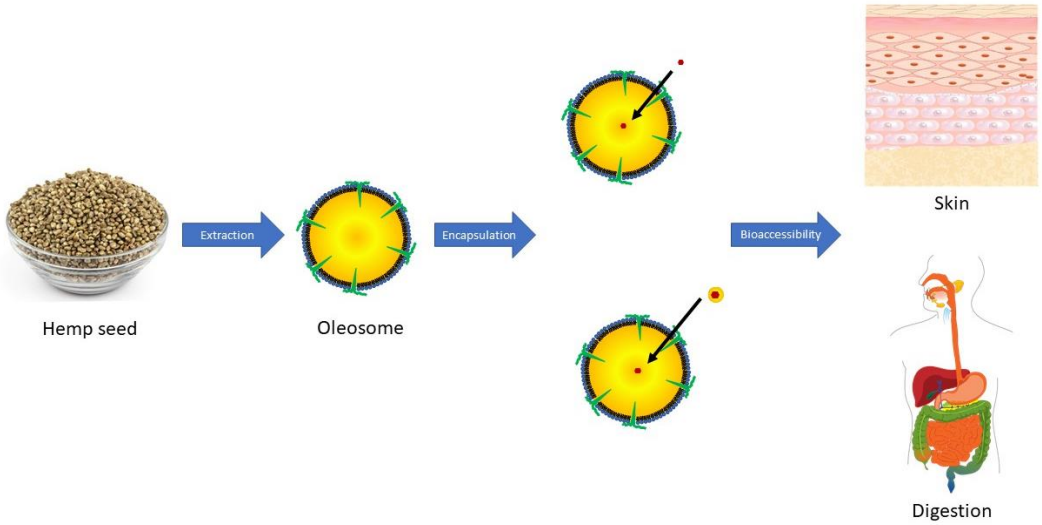
Increasing bioactivity of CBD by incorporating it in Lipid Droplets (Oleosomes)

Name PhD/PD: Zhaoxiang Ma
Involved staff members: Prof. dr. Costas Nikiforidis (BCT), Prof. dr. Harry Bitter (BCT), Prof. dr. Remko boom (FPE)
Project sponsor: Botaneco Inc. (Canada)
Start/(expected) end date of project: March 2020-February 2024

Background and goal of project

Lipid Droplets (Oleosomes) are natural plant-based oil droplets that have a triacylglycerol (TAG) core surrounded by a monolayer of phospholipids embedded with anchored proteins [1]. As a new type of natural delivery agent that can transport hydrophobic active molecules, like CBD, Lipid Droplets are a promising and challenging material. Compared with synthetic oil droplets, Lipid Droplets can be tailored made, are more sustainable and environmental-friendly which meet the consumer demands for healthy food and lifestyle.

This project aims to utilize hemp Lipid Droplets as carriers to transport hydrophobic functional ingredients for therapeutics. We will select the optimal Lipid Droplet extraction method, investigate the encapsulation of CBD in Lipid Droplets and determine the bioactivity of CBD, like the anti-inflammatory activity.



Highlight of the past year

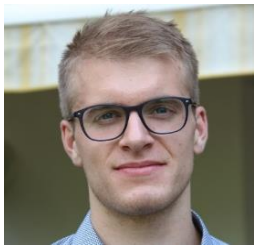
- Lipid Droplets were successfully extracted from hemp seeds.

Type of student projects envisioned

Student projects include lab work, focusing on encapsulation of CBD in Lipid Droplets and investigating the bioactivity of CBD in model human cells. For this research advanced analytical tools will be used, dynamic laser scattering and confocal microscopy.

1. Nikiforidis, C.V., *Structure and functions of oleosomes (oil bodies).* Advances in colloid and interface science, 2019: p. 102039.

Nature knows best – Lessons from plants on how to design stable emulsions resistant to lipid oxidation



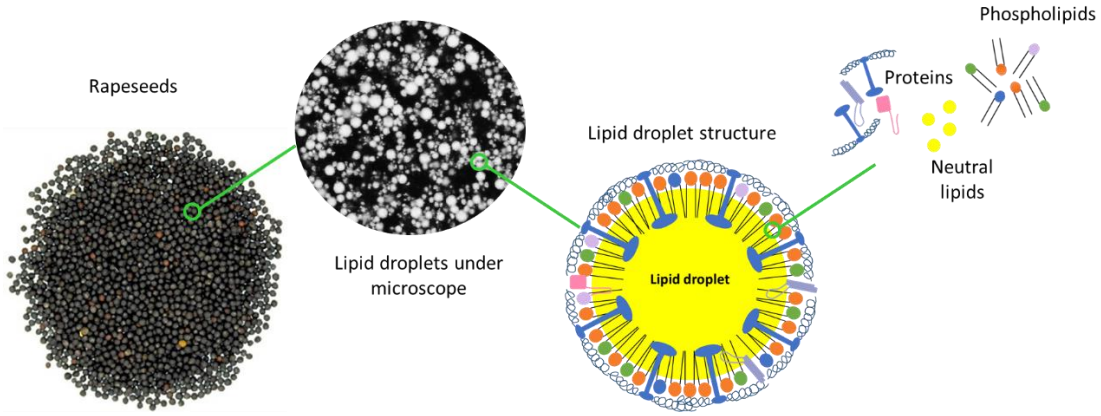
Name PhD/PD: Lorenz Plankensteiner
Involved staff members: Dr. Costas Nikiforidis (BCT), Dr. Marie Hennebelle (FCH), Prof.Dr. Jean-Paul Vincken (FCH)
Project sponsor: Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), Botaneco INC. (Calgary, AB, Canada)
Start/(expected) end date of project:
01.07.2020 / 01.07.2024

Background and goal of project

Preventing lipid oxidation of lipid-based products while meeting the increasing demand from consumers for natural and sustainable products; who says it is an impossible mission? It seems that plants already found the solution by storing their lipids in highly specialized and stable structures called Lipid Droplets (oleosomes), which consist of a core of neutral lipids stabilized by a combination of phospholipids and unique proteins. Now, we need to understand how it works. Lipid oxidation will be monitored in Lipid Droplets from different sources, with different composition and under different environmental conditions to characterize which properties confer them their oxidative resistance. The knowledge gathered will help broaden the field of applications for oleosomes, as a natural ingredient and develop nature-inspired emulsion systems.

Highlight of the past year

In my first year I focus on the extraction, purification and characterization of plant Lipid Droplets and their constituents. In the initial six months first important milestones were achieved. We were able to isolate pure Lipid Droplets from rapeseeds. Initial steps were taken to isolate and characterize the different building blocks of the Lipid Droplets. A new method was developed to isolate the unique protein of Lipid Droplets. The preliminary results obtained are very promising, higher protein amounts were isolated than with the methods previously reported in literature and a fairly high purity was achieved. This first achievements constitute a strong basis for further method development and optimisation.



Type of student projects envisioned

The unique approach in this project including extraction, chemical analysis and physical characterization of Lipid Droplets and their building blocks allows to offer a broad spectrum of thesis topics. Advanced analytical tools, laser scattering, tentiometry and microscopy will be used for the chemical and physicochemical analysis. The project can be tailored to the students interest.

Electrophoretic Lipid Droplet (Oleosome) and Protein Extraction from Oilseeds

Name PhD/PD: Kübra Ayan
Involved staff members: Dr. Costas Nikiforidis (BCT) & Prof. Remko Boom (FPE)
Project sponsor: National Education Ministry of Turkey
Start/(expected) end date of project: September 2020 / September 2024

Background and goal of the project

Oilseeds are valuable materials not only due to being the source of edible oil but also due to the high protein content. Oil (triacylglycerols) are stored in the seeds as Lipid Droplets (oleosomes), which contain oil in the core covered by a phospholipid – protein monolayer. Aqueous extraction of the Lipid Droplets results in a natural oil-in-water emulsion with high stability, which can be used in many applications, like foods to pharmaceuticals. Additionally, the oilseed proteins, like soy and rapeseed, can be used as an alternative to animal-based proteins because of growing protein demand[1,2]. However, lipid droplet and protein extraction are yet far from optimal, since it is energetically intensive and the use of chemicals is needed.

In the current study, we are developing a novel, single-step, and sustainable extraction process for Lipid Droplets and proteins by using electrophoretic techniques. We aim to obtain them simultaneously and reduce the use of water, organic solvents, and energy. Both Lipid Droplets and proteins are charged molecules so they can move under an applied electric field. This phenomenon is known as electrophoretic mobility and it depends on the electrical charge, size, shape of the molecules, and environmental conditions such as pH, salt concentration, and temperature[3]. We are expecting that Lipid Droplets and proteins will have different electrophoretic mobility and obtain them separately.

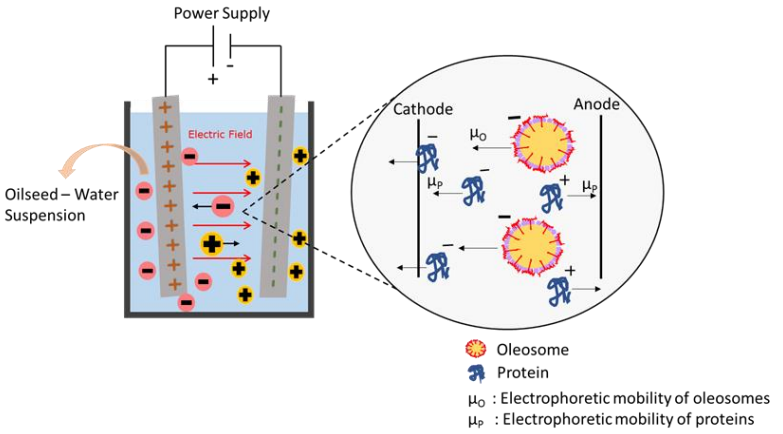


Figure 1: Schematic representation of electrophoretic extraction experimental set up.

Highlights for last year

This project started in September 2020. A highlight is that we measure the electrophoretic mobility of proteins and Lipid droplets from rapeseeds and they are significantly different.

Type of student projects envisioned

Potential student projects include lab works for the extraction of individual molecules, analysing their electrophoretic mobilities in specific conditions, and performing electrophoretic extraction.

1. Nikiforidis, C. V. (2019). Structure and functions of oleosomes (oil bodies). Advances in Colloid and Interface Science, 274, 102039. https://doi.org/10.1016/j.cis.2019.102039
2. Arrutia, F., Binner, E., Williams, P., & Waldron, K. W. (2020). Oilseeds beyond oil: Press cakes and meals supplying global protein requirements. Trends in Food Science & Technology. https://doi.org/10.1016/j.tifs.2020.03.044
3. Kurien, B. T., & Scofield, R. H. (Eds.). (2012). Protein electrophoresis: methods and protocols. Totowa, NJ: Humana Press.

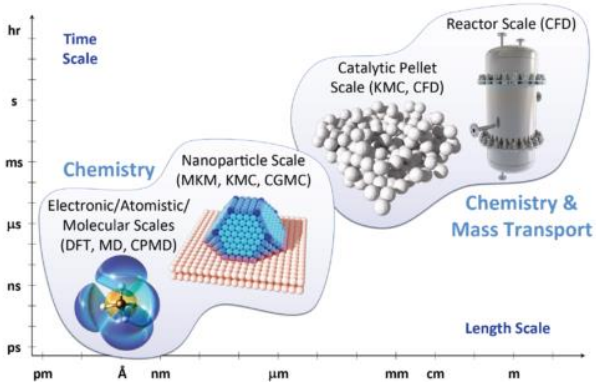
Modeling & Technology

Theme leader: Guanna Li (guanna.li@wur.nl)

Background and goal of this theme

Due to the fast development of computer architecture, nowadays we are able to carry out quantum simulations to address the structures of various heterogenous catalysts and the complex reaction networks of catalytic biomass conversion at atomic scale. On the other hand, the coupled flow, the heat and mass transfer, and reaction processes during biomass conversion can also be investigated by modeling at reactor scale. Such a combination of hierarchical approaches enable us to improve existing and design new catalyst and reactor technologies for a sustainable economy.

Therefore, in this theme we are aiming to unravel the fundament principles governing the experimentally observable reactivity and selectivity of different systems by multiscale modeling technologies.



Hierarchy of computational modeling

Main topics

- Solid (electro)catalysts for biomass conversion (e.g., starch oxidation)
  Development of kinetic models
  Investigation of diffusion in porous materials
- Capture of CO_2 from air (Direct Air Capture, DAC)
  Development of kinetic models
  Modeling condensation in micro- and mesopores
  Reactor/process design and heat integration
- Electro-conversion of biobased feeds into valuable platform chemicals
  Development of model electrocatalyst for biomass conversion
  Study of the effect of catalyst particle size on catalyst performance
- Multiscale modeling of supported solid catalyst for biomass conversion
  Investigation of the structure-reactivity relationships of metal carbide catalysts
  Development of operando modeling approaches

Student projects envisioned

Thesis subjects are related to the research work of PhD-students and Postdocs in the Modeling & Technology theme or in cooperation to the other BCT themes.

If you are interested in a thesis, please contact guanna.li@wur.nl to discuss specific details and possibilities.

Spuramolecular Continueous Flow Catalysis

Name PhD/PD: Tunan Gao
Involved staff members: Guanna Li, Fedor Miloserdov, Han Zuilhof, Harry Bitter
Project sponsor: VLAG graduate school
Start/(expected) end date of project: 12/2019-12/2023

Background and goal of project

Homogeneous catalysts are capable to mediate a number of unique transformations, that cannot be achieved by the conventional heterogeneous catalysts. However, the intrinsic drawbacks of separation and recycling difficulties often block their industrial application. In order to develop a catalysis system that combines the advantages of both homogeneous catalysis and heterogeneous catalysis, we propose to develop a novel flow catalytic system, that would benefit from the use of the unique structures of pillararenes as supramolecular hosts. First we are going to design a pillararene-based supramolecular host and a catalyst-linked guest that is strongly bound to host. Then the designed supramolecular host will be immobilized on a surface to capture the guest molecules with a homogeneous catalytic site on the tail. The non-covalent interaction between guest molecules (catalyst) and host molecules (pillararenes) will be used to prevent the catalyst from leaching. Such a semi-homogeneous catalyst is expected to be strongly bound to the macrocycles on the support. Because the non-covalent interaction is reversible, the deactivated catalyst can be removed and regenerated by specific stimulation treatments such as pH, salt, environment polarity, etc.

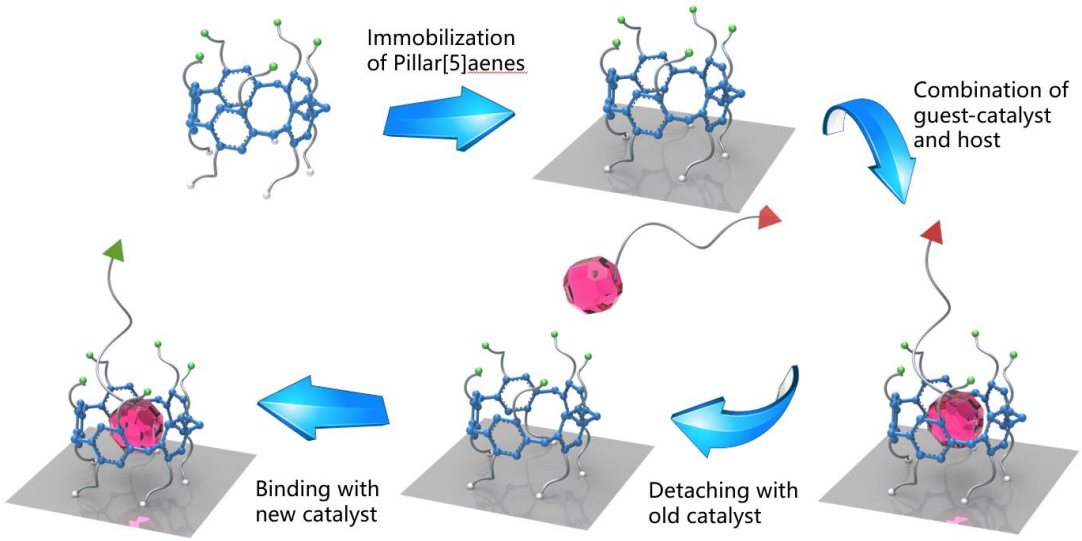


Figure 1. Schematic illustration of immobilization pillararenes on support and regeneration of deactivated catalysts with stimulations.

Highlight of the past year

We successfully synthesized the designed pillararenes in the past year. At this stage we are going to test the binding constant of host pillararenes and guest molecules. After that the pillararenes will be immobilized on a silica plate by click reactions and surface modification.

Type of student projects envisioned

This project mainly involves lab work, specifically organic synthesis skills. Characterization like NMR, MS, UV-vis will be applied. Also some surface chemistry and supramolecular chemistry techniques like NMR titration, surface modification, will be needed in this project.

Multiscale modeling of the supported solid catalysts for biomass conversion

Name PhD/PD: Raghavendra Meena, PhD candidate
Involved staff members: Prof. Harry Bitter, Prof. Han Zuilhof, Dr Guanna Li
Project sponsor: Sectorplan Chemistry & Physics
Start/(expected) end date of project: 01-10-2020/30-09-2024

Background and goal of the project
The depletion of fossil resources such as oil, coal and natural gas have initiated the search for renewable substitutes. Therefore, renewable resources such as biomass for chemicals and wind and solar for energy applications are emerging. The rapid development of the catalysis field coupled with the exponential increase in the computational resources motivates us to study the complex reactions involved in the biomass conversion. In our group, I model these catalysts and then try to understand such chemical reactions by simulating them on a supercomputer. I use the widely used periodic density functional theory (DFT) for performing these calculations.

Highlights of the past year
My project started a couple of months ago. I will use experimental data obtained by simultaneously running projects in our group (see e.g., Marlene Fuhrer). They provide me with the data of interesting catalysts, e.g., molybdenum carbides and tungsten carbides, which they found to be very efficient compared to the noble metal catalysts in the biomass valorization process. Now, I am studying these catalysts in detail. In particular, I am trying to uncover the property-performance relationship of these catalysts for the hydrodeoxygenation reaction of fatty acids.

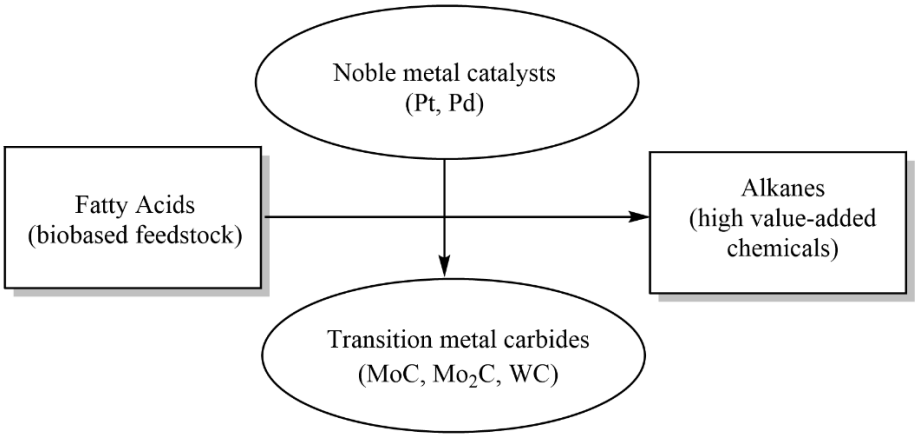


Figure 6: Scheme for biomass conversion. The horizontal flow shows the conversion of fatty acids into alkanes in the presence of a heterogeneous catalyst. The vertical flow indicates the substitution of noble metal catalysts with the transition metal carbide catalysts.

Type of student projects envisioned
I offer short term projects in which we will teach multiscale modeling. The nature of the project work can vary from understanding the density functional theory (DFT) to applying the knowledge of DFT and then performing calculations on a supercomputer.

Modelling of amino acids solubility in ethanol/water mixtures

Name PD: Dr. Carlos Cabrera
Involved staff members: Dr. Elinor Scott
Project sponsor: Greencovery
Start/(expected) end date of project: Nov 2019-Nov 2021

Background and goal of project

Commonly regarded as the building blocks of life, proteinogenic amino acids are valuable substances in many industries. They are used as food, as feed, in pharmaceuticals and they are a precursor for chemical ingredients used in a wide range of commercial products. Proteinogenic amino acids can be obtained from different sources, often classified by their three main production processes: chemical synthesis, fermentation and recovery from biomass. Common downstream processing consists of centrifugation, membrane filtration, chromatography, and crystallization. Two physiochemical properties especially of importance for designing chromatography and crystallisation are solubility and dissociation constants (pK's). In several, MSc projects the solubility of different amino acids were explored for ethanol/water mixtures from an experimental and modelling perspective.

Highlight of the past year

The solubility of amino acids in different ethanol-water mixtures varies because of solubility variation of the different ions (anion, cation, zwitterion) in the mixture and the variation of the dissociation constant in the different solvents. In recent projects, these two phenomena were studied separately from a modelling and experimental perspective. It was possible to explore the effect of each parameter separately and the result in the final solubility. Figure 1 shows the variation of solubility solubility for 8 different amino acids at different pH and ethanol composition.

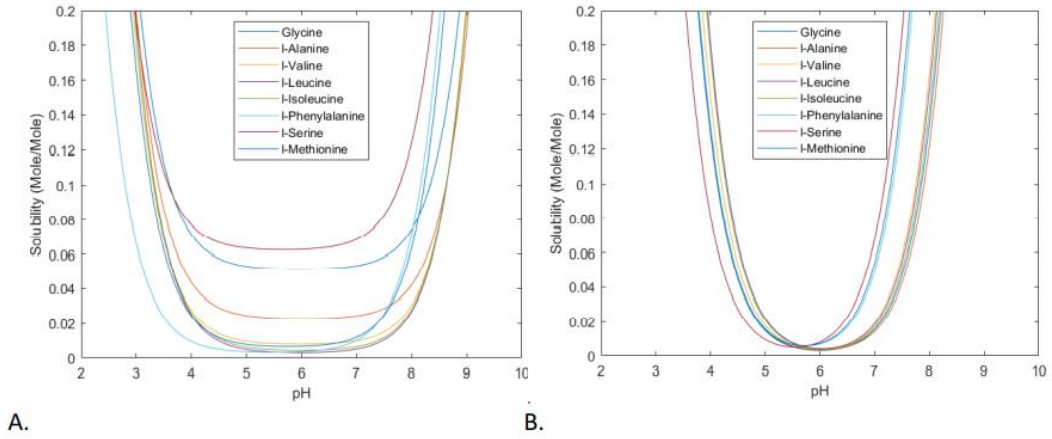


Figure 1. Solubility for 8 different amino acids at 100% water (1.A) and 20% water -80% Ethanol (1.B).

Type of student projects envisioned

Possible projects involved experimental and modelling of solubility of amino acids in Matlab and/or Aspen Plus.

Education

Bachelor and Master courses BCT and contribution to other courses

Code	Course title
BCT-20306	Modelling Dynamic Systems
BCT-22803	Physical Transport Phenomena
BCT-23306	Biorefinery
BCT-23806	Principles of Biobased Economy
BCT-24306	Renewable Resources and the (Bio)Chemical Production of Industrial Chemicals
BCT-30806	Physical Modelling
BCT-32306	Advanced Biorefinery
BCT-33806	Conversions in Biobased Sciences
BCT-34818	Micromaster Biobased Sciences for Sustainability

Code	Course title
BPE-10305	Process Engineering Basics
BPE-12806	Bioprocess Engineering Basics BT
BPE-60312	Bioprocess Design
FTE-12803	Introduction Biosystems Engineering part 2
ORC-12803	Organic Chemistry 1
ORC-12903	Organic Chemistry 2
ORC-13803	Bio-organic Chemistry for Life Sciences
PCC-33808	From Molecule to Designer Materials
YWU-60312	Research Master Cluster: Proposal Writing
YML-31304	Frontiers in Molecular Life Sciences

BCT: Biobased Chemistry and Technology
BPE: Bioprocess Engineering
FTE: Farm Technology
ORC: Organic Chemistry
PCC: Physical Chemistry and Soft Matter
YWU: Wageningen University
YML: Molecular Life Sciences

BSc Theses

- Imhof, Ruben; The effect of surface oxygen groups on the stability of platinum catalysts supported on carbon nanofibers for carbohydrate oxidation reactions
- Beelen, Bjorn; Physiochemical modification of spent graphite electrodes: Role of an additive
- Dam, Bastiaan van; Enzyme Kinetics of Production of Amylose from Sucrose
- Maas, Merijn; The influence of the temperature-programmed reaction, synthesizing carbon supported MoW-carbide catalysts, on the catalytic performance in stearic acid hydrodeoxygenation
- Dijkstra, Erwin; An 1H NMR analysis of the VCPO mediated conversion reaction of aspartic acid and β-methylaspartic acid

MSc Theses

- Cate, Evert ten; In vitro fermentation of linear resistant starch type 3 by the fecal microbiota of 3- and 7-week-old piglets
- Jolink, Steven; Modelling dissociation and solubility of 8 proteinogenic amino acids in ethanol-water mixtures
- Akerjiir, Stephanie; The use of heterogenous mix of aromatic molecules from lignin as medium in fermentation
- Costanzi, Nicola; Lignocellulosic biomass valorisation via molten salt and its economic assessment
- Reekum, Jasper van; Study of the physico-chemical behaviour of pea proteins as function of pH
- Feng, Rong; In vitro digestion and fermentation behavior of crystalline A- and B-type resistant starch III
- Vries, Jochem de; Monitoring the regeneration of cellulose within a lignocellulosic biorefinery
- Steijsiger, Remco; Characterisation of lignin isolated from wheat straw via a molten hydrated zinc chloride salt treatment
- Los, Sjoerd; Identification and characterisation of co-extracted lignin from wheat straw pretreated with a zinc chloride hydrate
- Verhagen, Simone; Catalytic hydrogenation of alginates derived from Alaria esculenta as a sustainable source for sugar alcohols
- Korte, Milan de; Investigating carbonation behaviour of K2CO3/AC during direct air capture
- Spoon, Martijn; Developing a kinetic model for the oxidation of polysaccharides over a heterogeneous platinum catalyst
- Shamusu Deen, Nafisah; Water condensation effect on carbon supported potassium carbonate sorbents in CO2 capture process
- Schwerdtfeger, Lorenzo; Towards Large Scale Biocatalytic Conversion of CO2 into Formic Acid using Industrial Flue Gas Streams
- Janssen, Fons; Modelling of CO2 utilization: energy savings via process integration of methanol synthesis with direct air capture
- Slaghek, Gillian; Modelling amino acid solubility in CO2 expanded ethanol

Scientific Publications 2020

Refereed article in a journal

- CO2 Conversion by Combining a Copper Electrocatalyst and Wild-type Microorganisms
  Chatzipanagiotou, Konstantina Roxani ; Jourdin, Ludovic ; Buisman, Cees J.N. ; Strik, David P.B.T.B. ; Bitter, Johannes H. (2020)
  ChemCatChem 12 (15). - p. 3900 - 3912.
- Assessment of rapeseed oil body (oleosome) lipolytic activity as an effective predictor of emulsion purity and stability
  Chirico, Simone De; Bari, Vincenzo di; Romero Guzmán, María Juliana ; Nikiforidis, Constantinos V. ; Foster, Tim ; Gray, David (2020)
  Food Chemistry 316 .
- Molybdenum and tungsten carbides can shine too
  Führer, M. ; Haasterecht, T. Van; Bitter, J.H. (2020)
  Catalysis Science & Technology 10 (18). - p. 6089 - 6097.
- Reaction Stages of Feather Hydrolysis : Factors That Influence Availability for Enzymatic Hydrolysis and Cystine Conservation during Thermal Pressure Hydrolysis
  Goerner-Hu, Xinhua ; Scott, Elinor L. ; Seeger, Thorsten ; Schneider, Oliver ; Bitter, Johannes H. (2020)
  Biotechnology and Bioprocess Engineering 25 (5). - p. 749 - 757.
- Ultrasonic inline inspection of a cement-based drinking water pipeline
  Hernandez Delgadillo, Hector ; Geelen, Caspar ; Kakes, Rutger ; Loendersloot, Richard ; Yntema, Doekle ; Tinga, Tiedo ; Akkerman, Remko (2020)
  Engineering Structures 210 .
- Assessment of air gap membrane distillation for milk concentration
  Moejes, S.N. ; Wonderen, G.J. van; Bitter, J.H. ; Boxtel, A.J.B. van (2020)
  Journal of Membrane Science 594 .
- Not sequentially but simultaneously : Facile extraction of proteins and oleosomes from oilseeds
  Ntone, Eleni ; Bitter, Johannes H. ; Nikiforidis, Constantinos V. (2020)
  Food Hydrocolloids 102 .
- Synthesis and characterization of a supported Pd complex on carbon nanofibers for the selective decarbonylation of stearic acid to 1-heptadecene : The importance of subnanometric Pd dispersion
  Ochoa, Elba ; Henao, Wilson ; Fuertes, Sara ; Torres, Daniel ; Haasterecht, Tomas Van; Scott, Elinor ; Bitter, Harry ; Suelves, Isabel ; Pinilla, Jose Luis (2020)
  Catalysis Science & Technology 10 (9). - p. 2970 - 2985.
- Influence of soaking time on the mechanical properties of rapeseed and their effect on oleosome extraction
  Romero-Guzmán, María Juliana ; Vardaka, Eirini ; Boom, Remko M. ; Nikiforidis, Constantinos V. (2020)
  Food and Bioproducts Processing 121 . - p. 230 - 237.
- Controlled oleosome extraction to produce a plant-based mayonnaise-like emulsion using solely rapeseed seeds
  Romero-Guzmán, María Juliana ; Köllmann, Nienke ; Zhang, Lu ; Boom, Remko M. ; Nikiforidis, Constantinos V. (2020)
  Food Science and Technology = Lebensmittel-Wissenschaft und Technologie 123 .
- Efficient single-step rapeseed oleosome extraction using twin-screw press
  Romero-Guzmán, M.J. ; Jung, L. ; Kyriakopoulou, K. ; Boom, R.M. ; Nikiforidis, C.V. (2020)
  Journal of Food Engineering 276 .
- The effect of monovalent (Na+, K+) and divalent (Ca2+, Mg2+) cations on rapeseed oleosome (oil body) extraction and stability at pH 7
  Romero-Guzmán, Maria Juliana ; Petris, Vasileios ; Chirico, Simone De; Bari, Vincenzo di; Gray, David ; Boom, Remko M. ; Nikiforidis, Constantinos V. (2020)
  Food Chemistry 306 .
- On the Emulsifying Properties of Self-Assembled Pea Protein Particles
  Sridharan, Simha ; Meinders, Marcel B.J. ; Bitter, Johannes H. ; Nikiforidis, Constantinos V. (2020)
  Langmuir 36 (41). - p. 12221 - 12229.

- Pea flour as stabilizer of oil-in-water emulsions : Protein purification unnecessary
  Sridharan, Simha ; Meinders, Marcel B.J. ; Bitter, Johannes H. ; Nikiforidis, Constantinos V. (2020)
  Food Hydrocolloids 101 .
- Harvesting time and biomass composition affect the economics of microalgae production
  Sui, Yixing ; Jiang, Yu ; Moretti, Michele ; Vlaeminck, Siegfried E. (2020)
  Journal of Cleaner Production 259 .
- Supported Cu Nanoparticles as Selective and Stable Catalysts for the Gas Phase Hydrogenation of 1,3-Butadiene in Alkene-Rich Feeds
  Totarella, Giorgio ; Beerthuis, Rolf ; Masoud, Nazila ; Louis, Catherine ; Delannoy, Laurent ; Jongh, Petra E. De (2020)
  The Journal of Physical Chemistry Part C: Nanomaterials and Interfaces 125 (1). - p. 366 - 375.
- Engineering the Protein Corona Structure on Gold Nanoclusters Enables Red-Shifted Emissions in the Second Near-infrared Window for Gastrointestinal Imaging
  Wang, Weili ; Kong, Yifei ; Jiang, Jun ; Xie, Qainqian ; Huang, Yang ; Li, Guanna ; Wu, Di ; Zheng, Huizhen ; Gao, Meng ; Xu, Shujuan ; Pan, Yanxia ; Li, Wei ; Ma, Ronglin ; Wu, Mei X. ; Li, Xuehua ; Zuilhof, H. ; Cai, Xiaoming ; Li, Ruibin (2020)
  Angewandte Chemie-International Edition 59 (50). - p. 22431 - 22435.
- Air-water interfacial and foaming properties of whey protein - sinapic acid mixtures
  Yang, Jack ; Lamochi Roozalipour, Sarah P. ; Berton-Carabin, Claire C. ; Nikiforidis, Constantinos V. ; Linden, Erik van der; Sagis, Leonard M.C. (2020)
  Food Hydrocolloids 112 .

Refereed book chapters

- Development perspectives for the bio-based economy
  Sanders, J.P.M. ; Langeveld, J.W.A. (2020)
  In: Biobased Products and Industries / Galanakis, C.M., Elsevier Inc., - p. 41 - 78.

PhD Theses

- Designing a sustainable oleosome aqueous extraction : A new way to make emulsion-based foods
  Romero Guzmán, María-Juliana (2020)
  Wageningen University. Promotor(en): R.M. Boom, co-promotor(en): C.V. Nikiforidis. - Wageningen : Wageningen University, - 183

Other output

Orals invited

Catalytic conversion of biomass - it is not only gold that glitters
Harry Bitter, VLAG, Wageningen, Netherlands, 17/6/2020

Shedding light on sorbent materials
Nazila Masoud, Dies Natalis 2020 WUR, Wageningen, Netherlands, 9/3/2020

From Msc to Prof.dr
Harry Bitter, Radboud University Nijmegen, Netherlands, 12/2/2020

Facile and efficient exploitation of plant materials
Costas Nikiforidis, University of Nottingham, United Kingdom, 3/2/2020

Challenges towards a sustainable future
Harry Bitter, Rotary Huissen, Netherlands, 28/1/2020

Biosourced polymers for designer materials
Costas Nikiforidis, China Agricultural University, China, 10/1/2020

Orals contributed

Boosting a biocathode by analysis: the invasive effects of cyclic voltammetry on microbial electrosynthesis
Sanne de Smit
International Chain Elongation Conference, Wageningen, Netherlands, 27/10/2020

Cyclic Voltammetry is invasive on Bioelectrochemical Systems
Sanne de Smit, Virtual Meeting of the International Society for Microbial Electrochemistry and Technologies, 8/10/2020

Carbon supported mixed metal-carbides for biomass conversion
Marlene Führer, International Conference on Environmental Catalysis (ICEC), Manchester, United Kingdom, 6/09/20 - 9/09/2020

Towards combining non-precious electro-catalysts and wild-type microorganisms for CO2 conversion
Roxani Chatzipanagiotou, International Conference on Environmental Catalysis (ICEC), Manchester, United Kingdom, 6/09/20 - 9/09/2020

Not sequentially but simultaneously: Facile extraction of proteins and oleosomes from oilseeds
Eleni Ntone, 15th International Hydrocolloids Conference, Melbourne, Australia, 2/03/2020 - 5/03/2020

The behaviour of sunflower oleosomes at the interfaces
Costas Nikiforidis, 15th International Hydrocolloids Conference, Melbourne, Australia, 2/03/2020 - 5/03/2020

Native pea protein aggregates as stabiliser of oil-in-water emulsions
Lakshminarasimhan Sridharan, 15th International Hydrocolloids Conference, Melbourne, Australia, 2/03/2020 - 5/03/2020

Steering site density in carbon supported of metal-carbides for deoxygenation of stearic acid
Marlene Führer, NCCC The Netherlands' Catalysis and Chemistry Conference (2020), Noordwijkerhout, Netherlands 2/03/2020 - 4/03/2020

Poster presentations

‘Cyclic Voltammetry is invasive on Bioelectrochemical Systems’
de Smit, S.M.; Buisman, C.J.N.; Bitter, J.H.; Strik, D.P.B.T.B., 71st annual meeting of International Society of Electrochemistry, Belgrade, Serbia, 31/08/2020 – 1/09/2020

‘Greener starch oxidation’
Hoogstad, T.M.; Buwalda, P.L.; Kiewidt, L.W.; Bitter, J.H., NCCC The Netherlands' Catalysis and Chemistry Conference (2020), Noordwijkerhout, Netherlands, 2/03/2020 - 4/03/2020

‘One step closer to combining electro-catalysis and microbial electro-synthesis’
Chatzipanagiotou, K.R.; Buisman, C.J.N.; Strik, D.P.B.T.B.; Bitter, J.H., NCCC The Netherlands' Catalysis and Chemistry Conference (2020), Noordwijkerhout, Netherlands, 2/03/2020 - 4/03/2020

Press/Media

An entrepreneur must be prepared to take risks
Wageningen World nr. 4 p. 40-43
https://edepot.wur.nl/536818
Carlos Cabrera Rodriguez
9/12/2020

New entrepreneurs need a helping hand
Wageningen World nr. 4 p. 44-45
https://edepot.wur.nl/536819
Carlos Cabrera Rodriguez
9/12/2020

Rennen op rubber en castorbonen hoe groen is deze schoen?
https://www.trouw.nl/duurzaamheid-natuur/rennen-op-rubber-en-castorbonen-hoe-groen-is-deze-schoen~be4973fc/
Trouw, 9/06/2020
Harry Bitter