



# abiosus e.V.

Supporting Research on Renewable Resources –  
For **a bio**-based and **sustainable** chemical industry

## 12<sup>th</sup> Workshop on Fats and Oils as Renewable Feedstock for the Chemical Industry

June 3 – 5, 2024  
Dortmund, Germany

in Cooperation with

Agency of Renewable Resources (FNR)  
TU Dortmund University

German Society for Fat Science (DGF)

German Chemical Society (GDCh), Division of Sustainable  
Chemistry



## Scientific and Organizing Committee

Ursula Biermann, abiosus e.V., Oldenburg, Germany

Jürgen O. Metzger, abiosus e.V. and University of Oldenburg, Oldenburg, Germany

Thomas Seidensticker, abiosus e.V. and TU Dortmund, Dortmund, Germany

Sebastian Kienast, Fachagentur Nachwachsende Rohstoffe e.V., Germany

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**MONDAY**  
**June 3, 2024**  
**Dorint, Picasso 1-3**

15:45		<b>Welcome &amp; Opening</b>	
16:00	L1	<b>Bornscheuer</b>	M
16:30	L2	<b>Katz</b>	M
17:00	L3	<b>Meier</b>	M
17:30		<b>Seidensticker</b>	
18:30		<b>Opening Mixer &amp; Poster Session</b> (18:30-21:30)	

**TUESDAY**  
**June 4, 2024**  
**Dorint, Picasso 1-3**

9:00	L4	<b>Kohl</b>	M
9:30	L5	<b>Nomura</b>	M
10:00	L6	<b>Grela</b>	D
10:20	L7	<b>Roth</b>	D
10:40		<b>Coffee Break</b>	
11:10	L8	<b>Hollmann</b>	M
11:40	L9	<b>Schörken</b>	M
12:10	L10	<b>Lorenzen</b>	D
12:30	L11	<b>Gröger</b>	D
12:50		<b>Lunch Break</b>	
14:00	L12	<b>Sturm</b>	M
14:30	L13	<b>Ramadas</b>	D
14:50	L14	<b>Cayli</b>	D
15:10	L15	<b>Di Serio</b>	D
15:30		<b>Coffee Break</b>	
16:00	L16	<b>Greiner</b>	M
16:30	L17	<b>Kleij</b>	M
17:00	L18	<b>Senthamarai</b>	D
17:20	L19	<b>Bizet</b>	D
19:00		<b>Conference Dinner</b>	

**WEDNESDAY**  
**June 05, 2024**  
**Dorint, Picasso 1-3**

9:00	L20	<b>Mutlu</b>	M
9:30	L21	<b>Harris</b>	D
9:50	L22	<b>Scheelje</b>	D
10:10	L23	<b>Gooßen</b>	D
10:30		<b>Coffee Break</b>	
11:00	L24	<b>Tilloy</b>	M
11:30	L25	<b>Leonori</b>	M
12:00	L26	<b>Häußler</b>	M
12:30		<b>Poster Award &amp; Closing Remarks</b>	
12:45		<b>End of Workshop</b>	
13:00		<b>Closing Lunch</b>	



# **Program**

## **Lectures and Posters**

**Monday, June 03, 2024**

**Registration**

*Registration will be open from 15:00 - 19:00*

**15.45 Welcome and Opening**

Thomas Seidensticker, abiosus e.V. and TU Dortmund

Markus Dierker, President of German Society of Fat Research (DGF)

**16.00 – 17.30 First Session**

*Chair: Thomas Seidensticker*

**16.00 – 16.30 Conversion of Fatty Acids Using Alpha-Dioxygenases and Engineered Lipxygenases to Valuable Compounds (M)**

L1 Uwe Bornscheuer, University of Greifswald, Greifswald, Germany

**16.30 – 17.00 Provision of Medium-Chain Fatty Acids (C12 - C18) from Local Circular Economy with Insect Biotechnology (M)**

L2 Heinrich Katz, Hermetia Baruth GmbH, Baruth, Germany

**17.00 – 17.30 Polyfunctional Renewable Monomers and (their Multicomponent Reaction Derived) Thermosets (M)**

L3 Michael A. R. Meier, KIT, Karlsruhe, Germany

**17.30 – 17.45 Introduction to Poster Session**

Thomas Seidensticker, TU Dortmund University, Dortmund, Germany

**18.30 – 21.30 Opening Mixer and Poster Session**

Posters will be displayed until the end of the workshop

(M) Main Lecture 30 min. including discussion

(D) Discussion Lecture 20 min including discussion



## Tuesday, June 04, 2024

9.00 – 10.30

*First morning session*

*Chair: Sébastien Tilloy*

9.00 – 9.30

**VerBioChem – Ethenolysis as a New Tool for the Chemical Industry (D)**

L4

Andreas Kohl, Verbio, Leipzig, Germany

9.30 – 10.00

**Biobased Chemically Recyclable Aliphatic Polyesters Prepared by Metathesis Polymerization (M)**

L5

Kotohiro Nomura, Tokyo Metropolitan University, Hachioji, Tokyo, Japan

10.00– 10.20

**Towards Practical Applications: Ethenolysis of Technical Methyl Oleate and FAME with Ruthenium Catalysts at Part-Per-Million Level (D)**

L6

Karol Grela, University of Warsaw, Warsaw, Poland

10.20 – 10.40

**For a World That's Bright, Use the Biodiesel Right, Don't Ignite – A Renewable Platform Chemical Enabled by Partial Hydrogenation (M)**

L7

Thomas Roth, Alexander Kühl, M. L. Spiekermann, H. W. Wegener, Dieter Vogt, Thomas Seidensticker, TU Dortmund University, Dortmund, Germany

10.40 – 11.10

*Coffee break*

11.10 – 12.50

*Second morning session*

*Chair: Uwe Bornscheuer*

11.10– 11.40

**Quo vadis Photodecarboxylase? (M)**

L8

Frank Hollmann, Delft University of Technology, Delft, The Netherlands

11.40 – 12.10

**Nylon-12 from Safflower Oil? A Biocatalytic Concept Study (M)**

L9

Anna Coenen, Valentin Gala Marti, Ulrich Schörken, TH Köln, Leverkusen, Germany

12.10 – 12.30

**Feedstock- and Product Diversification Technologies for Yeast Oil Production en-route to Commercialization (D)**

L10

Jan Lorenzen, Thomas Brück, Technical University of Munich, Garching, Germany

12.30 – 12.50	<b>Oleic Acid-Based Biodegradable Lubricants: Synthesis via Hydroformylation and Enzymatic Oligomerization &amp; Rheological properties (D)</b>
L11	Luisa Koch, <sup>1</sup> Alina Guntermann, <sup>1</sup> Katharina Hirschbichler, <sup>2</sup> Carmen Plass, <sup>1</sup> Tobias Betke, <sup>1</sup> Ling Ma, <sup>2</sup> Thomas Kiltthau, <sup>2</sup> <u>Harald Gröger</u> , <sup>1</sup> <sup>1</sup> Bielefeld University, Bielefeld, Germany; <sup>2</sup> Klüber Lubrication München GmbH & Co. KG, München, Germany
12.50 – 14.00	<i>Lunch break</i> <b>Restaurant Davidis</b>
14.00 – 15.30	<i>First afternoon session</i>  <i>Chair: Andreas Kohl</i>
14.00 – 14.30	<b>Exploring Bio-based Chemical Markets: Focus on Production and Feedstock Use in the EU (M)</b>
L12	<u>Viktorya Sturm</u> , Thünen-Institute of Market Analysis, Braunschweig, Germany
14.30 – 14.50	<b>Sustainability Without Performance Compromise: Novel surfactants solutions (D)</b>
L13	<u>Poorva Ramadas</u> , Clariant Produkte (Deutschland) GmbH, Burgkirchen, Germany
14.50 – 15.10	<b>Synthesis, Characterization, and Evaluation of Antibacterial Properties and Anion Removal Properties of Cationic Derivatives of Waste Cooking Oils (WCOs) (D)</b>
L14	<u>Gökhan Çayli</u> , Istanbul University-Cerrahpasa, Istanbul, Turkey
15.10 – 15.30	<b>Influence of Branching on the Physicochemical Properties of Saturated Fatty Acid Esters Synthesized from Linear and Branched Diols (D)</b>
L15	Michele Emanuele Fortunato, Rosa Vitiello, Vincenzo Russo, Francesco Taddeo, <u>Martino Di Serio</u> , University of Naples Federico II, Napoli, Italy
15.30 – 16.00	<i>Coffee break</i>

16.00 – 17.40	<i>Second afternoon session</i>
	<i>Chair: Mike Meier</i>
16.00 – 16.30	<b>Biobased Polycarbonates – Candidates for a Novel Class of Sustainable Engineering Plastics? (M)</b>
L16	<u>Andreas Greiner</u> , University of Bayreuth, Bayreuth, Germany
16.30 – 17.00	<b>Fatty Acid and Terpene Oxides as Versatile Precursors for Biobased Engineering Macromolecules (M)</b>
L17	Francesco Della Monica, <sup>1</sup> Arianna Brandolese, <sup>1</sup> Ilaria Grimaldi, <sup>1</sup> Lorenz Dittrich, <sup>1</sup> Carles Bo, <sup>1</sup> Alba Villar-Yanez, <sup>1</sup> Fernando Bravo, <sup>1</sup> <u>Arjan W. Kleij</u> , <sup>1,2</sup> <sup>1</sup> Institute of Chemical Research of Catalonia (ICIQ-Cerca), <sup>2</sup> Barcelona Institute of Science & Technology (BIST), Tarragona, Spain
17.00 – 17.20	<b>Terpene-Based Hybrid Polycarbonates via Co/Terpolymerization of Biobased Epoxides and CO: Access to Cross-Linked Polycarbonates with Enhanced Thermal Stability and Functionality (D)</b>
L18	<u>Thirusangumurugan Senthamarai</u> , <sup>1</sup> Arjan W. Kleij, <sup>1,2</sup> <sup>1</sup> Institute of Chemical Research of Catalonia (ICIQ), Tarragona, Spain; <sup>2</sup> Catalan Institute of Research and Advanced Studies (ICREA), Barcelona, Spain
17.20 – 17.40	<b>Vegetable Oil-Based Hyperbranched Materials – Synthesis and Derivatization Towards New Properties (D)</b>
L19	Guillaume Chollet, <sup>1</sup> Marie Reulier, <sup>1</sup> Etienne Grau, <sup>2</sup> Henri Cramail, <sup>2</sup> <u>Boris Bizet</u> , <sup>1</sup> <sup>1</sup> ITERG, CANEJAN, France ; <sup>2</sup> LCPO, Université de Bordeaux – CNRS – PESSAC, France
19.00	<i>Conference Dinner</i> <b>Restaurant Davidis</b>

## Wednesday, June 05, 2024

9.00 – 10.30      *First morning session*

*Chair: Arjan W. Kleij*

9.00 – 9.30      **New (Sustainable) Pathways to Integrate Sulfur Functional Groups into Renewable Based Polymers (M)**

L20      Cuong-Minh-Quoc Le, Adam Woodhouse, Yagmur Deniz Karatas, Bercis Pektas, Rayane Toutaoui, Gautier Schrodj, Hatice Mutlu, CNRS/Université de Haute Alsace, Mulhouse, France

9.30 – 9.50      **Optimisation of the RAFT Polymerisation of Pendant Fatty Acid Acrylamide Monomers Synthesised via the Base Catalysed Transesterification of Plant Oils (D)**

L21      Oliver Harris, Ryan Larder, Helen Willcock, Fiona Hatton, Loughborough University, Loughborough, United Kingdom

9.50 – 10.10      **Non-Isocyanate Polyurethanes from Terpene and Erythritol Feedstock using Organocatalysis and Thiol-Ene Reaction (D)**

L22      Clara Scheelje, Michael A. R. Meier, KIT, Karlsruhe, Germany

10.10 – 10.30      **Fatty Acid Valorization via Isomerizing Metathesis and Decarboxylative Ketonizations (D)**

L23      Lukas Gooßen, University of Bochum, Bochum, Germany

10.30 – 11.00      *Coffee Break*

11.00 – 12.30      *Second morning session*

*Chair: Andreas Greiner*

11.00 – 11.30      **Homogeneous Catalysis for the Functionalization of Oleochemicals and Their Derivatives into Value-Added Products (M)**

L24      Sébastien Tilloy, Université d'Artois, CNRS, Lens, France

11.30 – 12.00      **Photoexcited Nitroarenes as Ozone Surrogates (M)**

L25      Daniele Leonori, RWTH Aachen University, Aachen, Germany

12.00 – 12.30      **Fats and Oils - An Important Part of Transforming Chemistry to a Circular Economy (M)**

L26      Manuel Häußler, Max-Planck-Institute of Colloids and Interfaces (MPI-CI), Potsdam, Germany and Center for the Transformation of Chemistry (CTC)

12.30 – 12.45      **Best Poster Award and Closing Remarks**

Award committee: Azra Kocaarslan, Frank Hollmann, Ulrich Schörken

12.45      **End of Workshop**, followed by *Closing Lunch*, Restaurant Davidis

## Poster

- P1 **Continuous Selective FAME Hydrogenation Utilizing a Microcapillary Reactor**  
Florian Lehmann, Niclas von Vietinghoff, Takenobu Nakai, Lennard Nebel, Peter Pey, Karl Steffen Wulle, Dieter Vogt, Thomas Seidensticker, Laboratory of Industrial Chemistry, Department of Biochemical and Chemical Engineering, TU Dortmund University, Dortmund, Germany
- P2 **Biobased Linear/Network Aliphatic Polyesters Exhibiting Promising Tensile Properties**  
Kotohiro Nomura,<sup>a,\*</sup> Kosaku Tao,<sup>b</sup> Mika Kojima,<sup>a</sup> Xiuxiu Wang,<sup>a</sup> Lance O.P. Go,<sup>a</sup> Daisuke Shimoyama,<sup>a</sup> Mohamed M. Abdellatif,<sup>a</sup> Seiji Higashi,<sup>b</sup> and Hiroshi Hirano<sup>b\*</sup>, <sup>a</sup>Tokyo Metropolitan University, Hachioji, Tokyo, Japan; <sup>b</sup>Osaka Research Institute of Industrial Science and Technology (ORIST), Osaka, Japan
- P3 **New Cyclic(alkyl)(amino)carbene-Based Olefin Metathesis Catalysts for Green Chemical Conversion of Fatty Acids Derivatives**  
Filip Struzik, Biological and Chemical Research Centre, Faculty of Chemistry, University of Warsaw, Warsaw, Poland
- P4 **Conversion of Plant Oil Feedstocks *via* Continuous Flow Ethenolysis and Double Bond Isomerization**  
Lara N. Holderied, Inigo Göttker-Schnetmann and Stefan Mecking, Department of Chemistry, Chair of Chemical Material Science, University of Konstanz, Konstanz, Germany
- P5 **Synthesis of Biobased Poly(ester amide)s from Long-chain Fatty Acids and Oligopeptides**  
Gaku Miyamoto, Mohamed Mehawed Abdellatif, and Kotohiro Nomura\* Department of Chemistry, Tokyo Metropolitan University, Hachioji, Tokyo, Japan
- P6 **Biocatalytic Synthesis of *cis*-(+)-12-Oxophytodienoic Acid and Derivatization Using Metathesis**  
Maike Bittmann, Tim Lukas Guntelmann, Madita Knieper, Jana Löwe, Andrea Viehhauser, Harald Gröger, Karl-Josef Dietz, Bielefeld University, Bielefeld, Germany
- P7 **Production of Polyols from Linseed Oil by Reductive Hydroformylation**  
Walid Abdallah, Michel Ferreira, Hervé Bricout, Eric Monflier, Sébastien Tilloy, Univ. Artois, CNRS, Centrale Lille, Univ. Lille, Unité de Catalyse et Chimie du Solide (UCCS), Lens, France
- P8 **Poly(Menthene Carbonate): Synthesis and Depolymerization Studies**  
Enrico Lanaro<sup>a</sup> and Arjan W. Kleij<sup>a,b</sup>, <sup>a</sup> Institute of Chemical Research of Catalonia (ICIQ), the Barcelona Institute of Science & Technology (BIST), Tarragona, Spain; <sup>b</sup> Catalan Institute of Research and Advanced Studies (ICREA), Barcelona, Spain

- P9 **Catalyst-Free Synthesis of Bio-Derived Vitrimers Decorated with Dynamic Thioacetal Linkages**  
Yagmur Deniz Karatas, Cuong-Minh-Quoc Le, Gautier Schrodj and Hatice Mutlu,  
 Institut de Science des Matériaux de Mulhouse, CNRS/Université de Haute Alsace,  
 Mulhouse, France
- P10 **Project SymbioLoop: How to Convert Waste Fats and Oils into Fully Circular Plastics**  
Nico Friese,<sup>1</sup> Melissa De Rossi,<sup>1</sup> Patrick Rathenow,<sup>1</sup> Manuel Häußler<sup>1,2</sup>  
<sup>1</sup> Max-Planck-Institute of colloids and interfaces, Potsdam, Germany  
<sup>2</sup> Center for the Transformation of Chemistry (CTC), Delitzsch, Germany
- P11 **Sunflower Oil-Based Thermosets *via* the Passerini Three Component Reaction**  
Luis Santos Correa, Michael A. R. Meier, Institute of Biological and Chemical  
 Systems – Functional Molecular Systems (IBCS-FMS), Karlsruhe Institute of  
 Technology (KIT), Karlsruhe, Germany
- P12 **Diesel Fuel from Methyl Oleate by Sequential Double Bond Isomerization and Metathesis**  
Mykhailo Kondratiuk, Lukas Gooßen, Ruhr-Universität Bochum, Fakultät für Chemie  
 und Biochemie, Bochum, Germany.
- P13 **Novel Highly Efficient Catalysts for Ethenolysis**  
Rafał Gawin<sup>1</sup>, Andrzej Tracz<sup>1</sup>, Patryk Krajczyk<sup>1</sup>, Anna Kozakiewicz-Piekarz<sup>2</sup>, Juan  
 Pablo Martínez<sup>3</sup>, Bartosz Trzaskowski<sup>3</sup>, <sup>1</sup> Apeiron Synthesis SA, Wrocław, Poland  
<sup>2</sup> Faculty of Chemistry, Nicolaus Copernicus University in Toruń, Toruń, Poland,  
<sup>3</sup> Centre of New Technologies, University of Warsaw, Warszawa, Poland
- P14 **On the Degree of Unsaturation in the Pd-Catalyzed Alkoxy carbonylation of Oleochemicals – New Approaches and Insights**  
Hannes Wegener, Florian Lehmann, Thomas Seidensticker, Laboratory of  
 Industrial Chemistry, Department of Biochemical and Chemical Engineering, TU  
 Dortmund University, Dortmund, Germany
- P15 **C12-Polymer Precursors From Local Plant Oils Through Cross Metathesis and Isomerizing Hydroformylation**  
J. Hommes, D. Vogt, T. Seidensticker, Laboratory of Industrial Chemistry,  
 Department of Biochemical and Chemical Engineering, TU Dortmund University,  
 Dortmund, Germany
- P16 **From Fundamental Research to Start-up Founding**  
Maximilian Spiekermann, Max Krause, Jens Ehlhardt, Marén Schwandt, Thilo  
 Wattrodt, Thomas Seidensticker, Simplyfined, Laboratory of Industrial Chemistry,  
 Department of Biochemical and Chemical Engineering, TU Dortmund University,  
 Dortmund, Germany
- P17 **Bio-Polyols for High-Performance Composites From Local Vegetable Oils**  
Johannes Stiehm, Anika Hegemann, Harald Gröger, Bielefeld University, Bielefeld,  
 Germany

- P18 **Interreg VI A-Project 'Biotech Talent Unlocked' (31042)**  
Mark Rüschen gen. Klaas, Bernd Schmietenknop, University of Applied Sciences  
Emden/Leer, Emden, Germany
- P19 **Design and Development of Biodegradable Macromolecule Self-Assembly for Agriculture Formulations**  
Vinay Chauhan<sup>\*</sup>, Pankaj Sharma and Isha Soni, School of Advanced Chemical  
Sciences, Shoolini University, Solan, India
- P20 **Synthesis and characterization of PBT-Based Thermoplastic Copolyesters Containing a Renewable Fatty-Acid-Derived Soft Block**  
Apostolos A. Karanastasis<sup>1</sup>, Victoria Safin<sup>1</sup>, Subin Damodaran<sup>2</sup> and Louis M. Pitet<sup>1\*</sup>  
<sup>1</sup>Advanced Functional Polymers (AFP) Laboratory, Institute for Materials Research (IMO), Hasselt University, Hasselt, Belgium, <sup>2</sup>Tosoh Bioscience, GmbH, Griesheim, Germany
- P21 **Synthesis of Biobased and Biodegradable Polyurethane Coating from Olive Pomace Oil for Controlled-Release Fertilizer**  
Gökhan Çaylı<sup>1</sup>, Fatih Özönder<sup>2</sup>, <sup>1</sup> Istanbul University-Cerrahpaşa, Faculty of Engineering, Engineering Sciences, Istanbul, Turkey, <sup>2</sup> Istanbul Technical University, Department of Polymer Science and Technology, Istanbul, Turkey





# **Abstracts**

## **Part 1: Lectures**

## Conversion of Fatty Acids Using Alpha-Dioxygenases and Engineered Lipxygenases to Valuable Compounds

Uwe T. Bornscheuer

Department of Biotechnology and Enzyme Catalysis, Institute of Biochemistry, University of Greifswald, Greifswald, Germany  
uwe.bornscheuer@uni-greifswald.de

Enzymes are widely used for the modification of fats, oils and lipids ranging from applications in nutrition to oleochemistry.<sup>[1]</sup> Furthermore, natural enzymes can be tailor-designed using advanced methods of enzyme engineering to alter their substrate scope, selectivity, activity, thermostability, and solvent tolerance as described in our reviews.<sup>[2]</sup>

We have studied  $\alpha$ -dioxygenases ( $\alpha$ -DOXs) to convert fatty acids into valuable aldehydes.<sup>[3]</sup> Thus, capric acid and myristic acid could be efficiently converted into nonanal and tridecanal by oxidative decarboxylation using two  $\alpha$ -DOXs recombinantly expressed in *E. coli* originating from the cyanobacteria *Calothrix parietina* and LepDOX from *Leptolyngbya* sp.<sup>[4]</sup> In another project, we engineered a 13-lipoxygenase (13-LOX) originating from *Pseudomonas aeruginosa* (PaLOX), recombinantly expressed in *E. coli*, for the stereo- and regioselective C9-oxygenation of linoleic acid to produce (9S)-hydroxyoctadecenoic acid (9S-HODE). Two amino acids were identified as hot spots for switching the regioselectivity completely to C9-oxygenation (unpublished).

- [1] Biermann, U., Bornscheuer, U.T., Feussner, I., Meier, M.A.R., Metzger, J.O., *Angew. Chem. Int. Ed.* **2021**, 60, 20144-20165; Bornscheuer, U.T. (Ed.) *Lipid modification by enzymes and engineered microbes*, AOCS Press **2018**; Bornscheuer, U.T., *Ann. Rev. Food Sci. Technol.*, **2018**, 9, 85-103.
- [2] Buller, R., Lutz, S., Kazlauskas, R.J., Snajdrova, R., Moore, J.C., Bornscheuer, U.T., *Science*, **2023**, 382, eadh8615; Bornscheuer, U.T., Huisman, G., Kazlauskas, R.J., Lutz, S., Moore, J., Robins, K., *Nature*, **2012**, 485, 185-194.
- [3] Kim, I.J., Bayer, T., Terholsen, H., Bornscheuer, U.T., *ChemBioChem*, **2022**, 22, e202100693.
- [4] Kim, I.J., Brack, Y., Bayer, T., Bornscheuer, U.T., *Appl. Microb. Biotechnol.*, **2022**, 106, 197-210.

# **Provision of Medium-Chain Fatty Acids (C12 - C18) from Local Circular Economy With Insect Biotechnology**

Heinrich Katz

Hermetia Baruth GmbH

[h.katz@hermetia.de](mailto:h.katz@hermetia.de)

Insects are the animal class with by far the most species. Around one million species have been described. It is estimated that at least five million species have not yet been discovered. Thanks to their fantastic characteristics and abilities, insects colonize almost all habitats on earth. We humans can also benefit from the many survival strategies they have developed. Insect Biotechnology, the yellow biotechnology comprises the development of bioresources for use in medicine, agriculture (plant protection, pollination, feed), food and technology industry. Insects are an important link in the natural circular economy. Their main task in nature is to remove waste by converting it into higher-value products, such as protein and fat.

Hermetia Baruth GmbH (HBG) started in 2006 with the production of *Hermetia illucens* (Black Soldier Fly = BSF) and achieved the first stable and significant population in Europa. An overview of the production process is given. It is divided into substrate acquisition and preparation, insect rearing (holometabolic life cycle), insect fattening and harvesting process. Further separation of insect frass and live larvae as well as breaking down into the components proteins and lipids. The core product is the protein rich insect meal, but also the by-products lipid, fertilizer and Chitin have a value and support the viability of the approach. Especially the lipid can be used as base material for a variety of sustainable end products. The spectrum shows a high content of lauric acid (C12) and palmitic acid (C16). These medium chain fatty acids can be used to gain technical products like fuel (Diesel, Kerosene), lubricants, bio surfactants, as well as cosmetics (e.g. lip balsam, skin cream). A big advantage to use the insect lipids as a base for technical products is the chance to stay outside the feed and food chain. So, there would be no risk in terms of food safety. However, the European legislation does not support this. There are strong movements going on to change this and support the circular economy approach with insects. Nature does not know any waste, so let's use insects according to their capabilities and natural behavior!

## Polyfunctional Renewable Monomers and (Their Multicomponent Reaction Derived) Thermosets

Michael A. R. Meier

Institute of Organic Chemistry (IOC) and Institute of Biological and Chemical Systems –  
Functional Molecular Systems (IBCS-FMS), Karlsruhe Institute of Technology (KIT),  
Karlsruhe, Germany  
m.a.r.meier@kit.edu

Multifunctional monomers are important starting materials for a range of cross-linked materials, including, for instance, polyurethanes or epoxy resins. The development of renewable multifunctional monomers was traditionally largely focused on polyols for polyurethane synthesis. Here, approaches to new monomers based on renewable resources, mainly from plant oils and lignin, will be discussed with a focus on (but not limited to) using new cross-linking chemistry based on multicomponent reactions. Exemplarily, plant oil derived resins and their adhesive performance based on the Passerini three component reaction will be discussed. Moreover, the catalytic activation of elemental sulfur for the *in situ* synthesis of isothiocyanates will be introduced, which can, again *in situ*, be cross-linked with polyols in an overall three component reaction. Also other approaches towards cross-linked materials based in lignin derived monomers, based on the reaction of lignin with organic carbonates, will be discussed.

## **VerBioChem – Ethenolysis as a New Tool for the Chemical Industry**

Andreas Kohl

Verbio SE, Ritterstraße 23, 04109 Leipzig  
Andreas.Kohl@verbio.de

Reacting to the growing market needs in the chemical industry for biobased, renewable and chemicals with a low CO<sub>2</sub> footprint, Verbio SE has developed a process to produce methyl-9-decenoate, 1-decene and C18 diacids derivatives from renewable rapeseed methyl ester by olefin metathesis / ethenolysis. Olefin metathesis is a versatile reaction to generate unique molecules especially useful if used in combination with renewable oleochemical feedstock.

The new process VerBioChem, provides access to these medium chain alpha olefins in an environmentally friendly and economically attractive way from readily available rape seed methyl ester. Furthermore, the metathesis platform can be used to produce a number of useful C18 diacids e. g. dimethyl 9-octadecenedioate or octadecanedioate by self-metathesis as new biobased building blocks for the chemical and especially the polymer industry.

Verbio is in the process of building an ethenolysis production plant with 60 kta product capacity in Germany as well as a commercial scale metathesis catalyst production facility in Hungary.

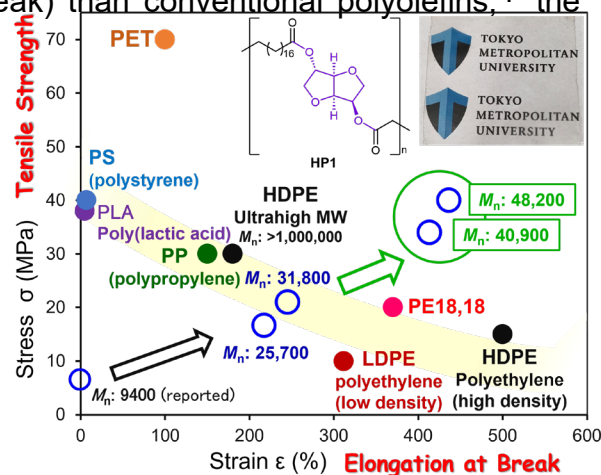
Here we will provide insight into the ethenolysis technology, the renewable products spectrum, as well as give an update on the construction of the ethenolysis and catalyst production plant. This investment shows Verbio's believe that especially XiMo Hungary kft's Schrock type metathesis catalysts provide a commercially attractive opportunity and give access to new commercially relevant reaction space for the Chemical industry.

## Biobased Chemically Recyclable Aliphatic Polyesters Prepared by Metathesis Polymerization

Kotohiro Nomura

Tokyo Metropolitan University, Hachioji, Tokyo, Japan  
ktnomura@tmu.ac.jp

This paper focused on synthesis of biobased aliphatic polyesters by acyclic diene metathesis (ADMET) polymerization and the subsequent hydrogenation,<sup>1-3</sup> in particular synthesis of high molecular weight polyesters by adopting the polymerization using molybdenum-alkylidene catalysts. The resultant polymer film exhibit better tensile properties (tensile strength and elongation break) than conventional polyolefins;<sup>4</sup> the network polymers, prepared in the presence of glycerol based cross linkers, improved the elongation at break.<sup>5</sup> Moreover, the resultant polyesters including conventional polyesters (PET, PBT etc.) could be converted to monomers quantitatively by transesterification with alcohol in the presence of  $\text{Cp}^*\text{TiCl}_3$  ( $\text{Cp}^* = \text{Cp}, \text{C}_5\text{Me}_5$ ).<sup>6</sup>



In this symposium, we wish to present our results including our update data concerning synthesis new polyesters, poly(ester amide)s showing better tensile properties including relationship with the monomer repeating units. Detail will be introduced in the symposium.

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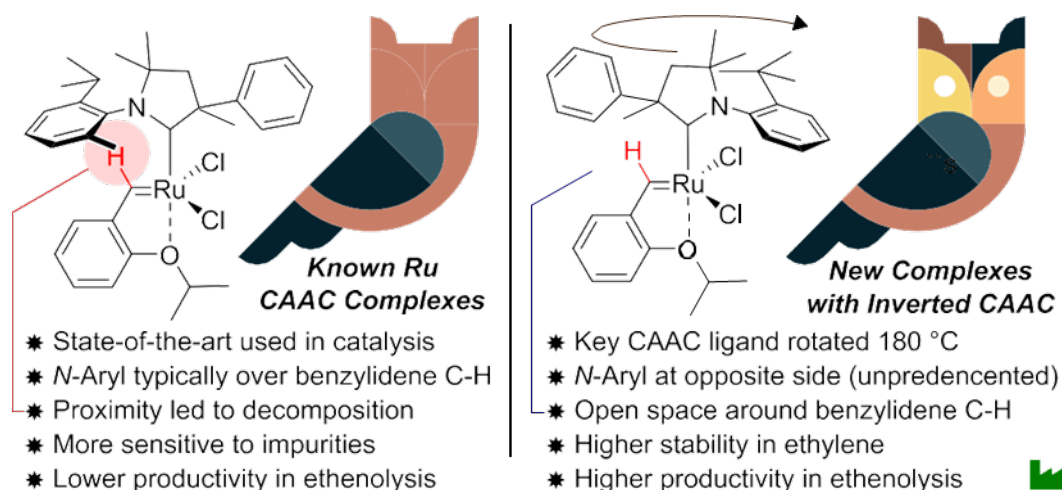
## Towards Practical Applications: Ethenolysis of Technical Methyl Oleate and FAME with Ruthenium Catalysts at Part-Per-Million Level

Karol Grela

Biological and Chemical Research Centre, Faculty of Chemistry, University of Warsaw,  
Żwirki i Wigury Street 101, 02-089 Warsaw, Poland prof.grela@gmail.com

www.karolgrela.eu

Metathesis opened up potential opportunities to produce numerous fine chemicals from a biomass. From many types of olefin metathesis, the ethenolysis (cross metathesis with ethylene) can be used to convert many plant-derived raw materials into valuable fine chemicals. One of potential substrates are plant and algae oils, rich in oleic acid. Use of oleic acid esters in ethenolysis were however hampered by the lack of air- and moisture-stable and productive catalysts for this reaction. During the lecture, some new and highly potent Ru-catalysts systems for ethenolysis of a biomass will be presented that work in air and with technical-grade biomass substrates (FAME).<sup>1-2</sup>



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## For a World That's Bright, Use the Biodiesel Right, Don't Ignite – A Renewable Platform Chemical Enabled by Partial Hydrogenation

Thomas Roth, Alexander Kühl, Maximilian. L. Spiekermann, Hannes. W. Wegener,  
Dieter Vogt, Thomas Seidensticker

TU Dortmund University, Department for Biochemical and Chemical Engineering,  
Laboratory of Industrial Chemistry, Emil-Figge-Straße 66, 44227 Dortmund/Germany  
Email: thomas2.roth@tu-dortmund.de

The hydrogenation of polyunsaturated fatty acids (PUFAs) in vegetable oils and their derivatives is essential for their use in many areas, such as biofuels and food chemistry. However, no attempts have been made to adapt this technology to the requirements of further chemical utilisation of fatty acid methyl esters as molecular building blocks, especially for the particularly promising functionalisation of the C=C double-bond. In this work, we, therefore, use three homogeneously catalysed model reactions (hydroformylation, isomerising methoxycarbonylation, and ethenolysis) to demonstrate that high PUFA contents have a negative impact on activity and selectivity, as already shown in various literature.<sup>[1,2]</sup> Subsequently, using the example of soybean and canola biodiesel, we demonstrate that applying partially hydrogenated biodiesel with a PUFA share of <1 w% significantly increases activity (up to twentyfold) and selectivity (up to 80% increase) in these reactions. In a further study, the possible reasons for the observed catalyst deactivation were investigated. Using hydroformylation as an example, the negative influence of PUFAs on the catalytic conversion of methyl oleate was precisely demonstrated. The mechanism of this phenomenon was described and demonstrated using several analytical methods. This research provides the framework for increasing the industrial use of renewable raw materials such as biodiesel and thus guiding our value chain towards a sustainable future.

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## Quo Vadis Photodecarboxylase?

Frank Hollmann

Department of Biotechnology, Delft University of Technology

f.hollmann@tudelft.nl

Since their discovery in 2017, Fatty Acid Photodecarboxylases (FAPs)<sup>1</sup> have been receiving tremendous attention in the biocatalysis community. FAPs catalyse the light-driven decarboxylation of a broad range of carboxylic acids into the corresponding alkanes. The thermodynamic driving force needed for this reaction is obtained from blue light absorbed by the flavin cofactor.



A particular interest in FAPs derives from the vision of producing next generation biofuels (i.e. alkanes instead of fatty acid methyl esters) from waste fatty acids.

FAPs, however, also suffer from a pronounced instability against blue light thereby challenging the economic feasibility of FAPs for fuel applications.

In this presentation I will present the current state-of-the-art of FAP-discovery and -engineering and preparative applications. A particular focus will be on the photoinactivation of FAPs and some possible solutions.

1. Sorigué, D.; Légeret, B.; Cuiné, S.; Blangy, S.; Moulin, S.; Billon, E.; Richaud, P.; Brugière, S.; Couté, Y.; Nurizzo, D.; Müller, P.; Brettel, K.; Pignol, D.; Arnoux, P.; Li-Beisson, Y.; Peltier, G.; Beisson, F. An algal photoenzyme converts fatty acids to hydrocarbons, *Science* 2017, 357, 903-907.

## Nylon-12 From Safflower Oil? - A Biocatalytic Concept Study

Anna Coenen, Valentin Gala Marti, Ulrich Schörken

TH Köln – Campus Leverkusen, Campusplatz 1, 51379 Leverkusen, Germany

[ulrich.schoerken@th-koeln.de](mailto:ulrich.schoerken@th-koeln.de)

13-specific lipoxygenases (LOX) and hydroperoxide lyases (HPL) catalyze the consecutive hydroperoxidation of linoleic acid to the 13-hydroperoxide (13-HPODE) followed by splitting to volatile hexanal and 12-oxododecenoic acid. Hexanal is marketed by the flavor and fragrance industry as green note. In contrast, the bifunctional oxoacid is not used industrially up to now, though it could be an interesting bio-based polymer precursor for C12  $\omega$ -hydroxy acid, dicarboxylic acid or  $\omega$ -amino acid synthesis.

Here we present the development of an enzyme cascade starting from safflower oil to produce 13-HPODE in high yield. Lipase was coupled to the LOX reaction and *in situ* oxygen production was achieved with catalase under continuous hydrogen peroxide dosage. The utilization of green surfactant Triton CG-110 improved the reaction significantly. All three enzymes were additionally immobilized to evaluate their use in repeated batches.

Next, we cloned four HPLs and evaluated for their 13-HPODE splitting potential. Best performance was obtained upon N-terminal truncation and HPL from papaya was selected as best performing enzyme. Cascade synthesis of 12-oxododecenoic acid was achieved by coupling of HPL with LOX and lipase. Finally, seven transaminases ( $\omega$ -TAs) were expressed in *E. coli* and tested for their 12-oxododecenoic acid transformation potential. All enzymes accepted the substrate and the successful formation of aminododecenoic acid was proven analytically. In an enzyme cascade with LOX, HPL and  $\omega$ -TA were able to synthesize 12-aminododecenoic acid from linoleic acid in a one-pot reaction.

## Feedstock- and Product Diversification Technologies for Yeast Oil Production en-route to Commercialisation

Jan Lorenzen, Thomas Brück

Werner Siemens Chair for Synthetic Biotechnology, School of Natural Sciences, Dept. of Chemistry, Technical University of Munich, Garching, Germany

Email: jan.lorenzen@tum.de

Over the last 12 years our research group has developed a comprehensive, new production platform for yeast oil generation, using the non-conventional yeast *Cutaneotrichosporon oleanum* as a production organism. This production platform differentiates itself from conventional yeast oil processes, as it operates with a monoauxic process, that allows simultaneous generation of yeast biomass and triglycerides with high space-time and total yields. The unit operations of the process from raw material to oil product constitute yeast **Fermentation** using complex carbon containing feedstocks, enzymatic **Hydrolysis** of the yeast cell wall, **Centrifugation** to recover the triglyceride based yeast oil and finally **Recycle** all aqueous side stream streams in upstream unit operations. Hence we named this holistically patented process FHCR™, as it does not generate waste streams or residue water, all while saving on upstream feedstock costs as non-oil cell residues are recycled in the next round of fermentation. Specifically, the intracellular triglycerides are recovered by a tailored enzymatic process followed by a one step centrifugation step providing a clean oil fraction, which in contrast to plant oils and other yeast oil products does not require further purification prior to applications in the food, chemical or cosmetic sector. This talk will report on recent developments regarding the diversification of the feedstock base for yeast oil production focusing on complex biomass waste streams from pulp and paper and citric acid processing as well as the valorization of yeast oil contained secondary metabolites, specifically squalene and steroids. Further, the enzymatic upgrading of yeast oil derived fatty acids using cell-free enzyme cascade will be discussed. One of the flagship research activities synchronizing all these technology advances is “Valuable” EU Project lead to our research group.

## **Oleic Acid-Based Biodegradable Lubricants: Synthesis *via* Hydroformylation and Enzymatic Oligomerization & Rheological Properties**

Luisa Koch,<sup>1</sup> Alina Guntermann,<sup>1</sup> Katharina Hirschbichler,<sup>2</sup> Carmen Plass,<sup>1</sup> Tobias Betke,<sup>1</sup>

Ling Ma,<sup>2</sup> Thomas Kilthau,<sup>2</sup> Harald Gröger<sup>1</sup>

<sup>1</sup> Bielefeld University, Bielefeld, Germany

<sup>2</sup> Klüber Lubrication München GmbH & Co. KG, München, Germany

harald.groeger@uni-bielefeld.de

The development of bio-based and biodegradable lubricants is a research area of high current interest. In this contribution, we present the results of a joint academic-industrial collaboration project addressing the development of a new generation of bio-based lubricant materials.[1] The raw material basis is oleic acid as a readily available and economically attractive unsaturated fatty acid, which is accessible, e.g., by means of high oleic sunflower oil. The synthetic access to the desired lubricant molecules, which show an oligomeric structure, proceeds through a hydroformylation of methyl oleate, followed by hydrogenation and a subsequent solvent-free enzymatic oligomerization *via* transesterification. This biocatalytic steps runs in a solvent-free mode by means of an immobilized lipase as a heterogeneous catalyst. The developed synthetic process allows an economic production of the lubricant products. A specific feature of this new generation of oligomeric lubricant molecules is their modularity. For example, the chain length can be adjusted in a tailor-made form according to the needed rheological properties such as, e.g., pour point or viscosity.[1] Furthermore, the developed lubricant products have been experimentally proven to be biodegradable in an OECD 301 F test.

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## Exploring Bio-Based Chemical Markets: Focus on Production and Feedstock Use in the EU

Viktoriya Sturm

Thünen Institute of Market Analysis, Braunschweig, Germany

viktoriya.sturm@thuenen.de

The production of bio-based chemicals is driven by the need to reduce industry's dependence on non-renewable fossil-based raw materials. Although the production of bio-based chemicals is associated with positive effects, such as the reduction of GHGs emissions, increased employment and the introduction of products with new functionalities, risks are also being discussed. The most prominent are related to the sustainable use of biomass as feedstock: How much bio-based production could be realized without triggering the dilemma of trade-offs for the use of biomass (food-feed-energy-materials)? Which bio-based chemicals are the main contributors to the overall industrial demand for biomass? Which feedstock use needs special attention?

Answers to this question are of importance for policymakers who are framing the playing field for producers but the industry itself is also interested. Although a number of efforts to establish the bioeconomy monitoring system on national and EU level have already been undertaken, current available data provides very limited insights into the national/ EU bio-based chemical industry. We have addressed this shortcoming by developing the **BioMAT** (**Bio**-based **MAT**erials), a tool which comprises two elements: a comprehensive database (BioMAT database) and a multi-regional partial equilibrium model (BioMAT model). While the BioMAT database tracks historical developments in the markets for bio-based chemicals and the associated demand for bio-based feedstocks, the BioMAT model uses this information and projects their future developments. BioMAT is therefore an appropriate tool for showing prospects of the bio-based industry, but also revealing its limitations and the associated trade-offs.

## **Sustainability Without Performance Compromise: Novel Surfactants Solutions**

Poorva Ramadas

Clariant Produkte (Deutschland) GmbH, Burgkirchen, Germany

Poorva.ramadas@clariant.com

As a leading company in the field of specialty chemicals, Clariant is committed to the development of innovative and sustainable surfactant solutions for our customers, without compromising on the performance. Glucamides are special oleochemicals-based amides which are derived from the renewable feedstocks glucose, fatty acids and / or esters. In addition to being sustainable, glucamides offer unique performance profiles, which are better than many current fossil and bio-based market solutions. In this presentation, two Glucamides will be covered, which address both sustainability and performance requirements of our customers for various applications:

- 1) Velsan Flex: An innovative, bio-based and multifunctional preservation booster based on a cyclic glucamide surfactant, which is highly water-soluble and pH stable allowing broad formulation flexibility in Personal Care applications.
- 2) GlucoPure Sense: an ultra-low product carbon footprint glucamide surfactant based on high oleic sunflower oil which offers a creamier foam and better sensory feel on skin and excellent cleansing properties, allowing it to be used-in rinse-off formulations and hand dish-washing liquids.

## **Synthesis, Characterization and Evaluation of Antibacterial Properties and Anion Removal Properties of Cationic Derivatives of Waste Cooking Oils (WCO's)**

Gökhan Çaylı

Istanbul University-Cerrahpasa, Istanbul, Turkey

gokhan.cayli@iuc.edu.tr

The amount of waste cooking oils (WCO's) increases linearly with global population. The current estimate of the world's population is 7.79 billion, and according to projections, between 12 and 13 billion people will inhabit the planet by the year 2100. It is apparent that as the population grows, the amount of used cooking oil would increase as well. The bulk of used cooking oil is produced in the United States and the People's Republic of China. Only a few sources of pollution exist in the rest of the world, such as those found in the countries of the European Union. A crucial concern would be recycling those materials. Two important issues on WCO that scientists are studying are how and in which ways waste cooking oils will be utilized. First solution of those issues is to refine and reuse of the waste cooking oil by special processes and additives. It is not safe reuse waste cooking oil for food purposes. Thus, it is necessary to find WCO's alternative application areas. For the safe applications, the structure of WCO's should be modified. Although cooking process may cause some minor changes in their structures, WCO can give almost all the reactions of a typical triglycerides.

In this study, epoxy derivative of waste cooking oil synthesized in the first step. Then EWO was reacted with monochloroacetic acid. When this chlorinated derivative is reacted with tertiary amines, quaternarization take place. Triethylamine and pyridine were used for modification. Those quaternary ammonium salts are water soluble and have anti-bacterial potential and they exhibit good anion removal properties.

## Influence of Branching on the Physicochemical Properties of Saturated Fatty Acid Esters Synthesized From Linear and Branched Diols

Michele E. Fortunato, Rosa Vitiello, Vincenzo Russo, Francesco Taddeo, Martino Di Serio

University of Naples Federico II, Department of Chemical Science

Fatty acid alkyl esters (FAAE) are increasingly gaining popularity as bio-based lubricants due to their superior lubricity compared to mineral-based oils and biodegradability [1,2]. The physical-chemical and rheological properties of FAAE are strictly dependent on the chemical structure of the carboxylic acid and the alcohol from which they are derived [3]. Oleic acid esters are usually employed in the formulation of bio-based lubricants, but the presence of unsaturation dramatically decreases its oxidation stability. On the other hand, saturated fatty acid esters demonstrate a notable advantage in terms of oxidative stability compared to their unsaturated counterparts, enhancing the overall performance and longevity of bio-based lubricants but they present poor low temperature properties.

This research work focuses on the synthesis of esters derived from saturated fatty acids ( $C_9$ ,  $C_{12}$ , and  $C_{14}$ ) and linear or branched diols ( $C_2$ - $C_4$ ). The primary objective is to evaluate the impact of branching on the physicochemical properties of the final product. The esters were synthesized with a purity level at least of 98 wt%, as verified by H-NMR analysis. Notably, the transition from esters derived from linear diols to their branched counterparts results in a significant reduction in pour point. As an example,  $C_{12}$  esters obtained from 1,3-propanediol are solid state at room temperature, while those derived from 2,3-propanediol (branched form) appears liquid. This distinct effect persists consistently up to  $C_{12}$  esters, with  $C_{14}$  esters remaining solid due to the prevailing influence of molecular weight effects over the impact of branching degree. These obtained results can be a useful tool for the development of bio-based lubricants tailored for low-temperature applications and hold potential as valuable additives in bio-based lubricant formulations.

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## **Biobased Polycarbonates – Candidates for a Novel Class of Sustainable Engineering Plastics?**

Andreas Greiner

Macromolecular Chemistry and Bavarian Polymer Institute, Universität Bayreuth,  
Universitätsstraße 30, 95440 Bayreuth, Germany

Biobased polymers are a promising alternative to fossil-based polymers. They offer access to unique physical and chemical properties, making them particularly interesting. The copolymerization of trans-limonene epoxide and CO<sub>2</sub>, catalyzed by organo-zinc, produces poly(limonene carbonate) (PLimC) with exceptional physical and chemical properties. PLimC yields highly transparent coatings, is scratch-resistant, and exhibits good selectivity in gas permeation. Additionally, it has a high molecular weight and a glass transition temperature of 135°C. The lateral double bond in each repeating unit of PLimC makes it a perfect platform for functional polymers. These double bonds allow for a wide variety of chemical modifications, resulting in polymers with novel properties. PLimC exhibits unique permeation properties and excellent transparency, allowing for the creation of a new class of materials known as 'breathing glass'. The high permeability of CO<sub>2</sub> enables exchange with the surrounding environment through a closed PLimC window, without the need to open the window.

The limitations of melt processing of PLimC were successfully overcome by adding biobased softeners that do not leach out. PLimC offers great potential as an engineering plastic. However, there are still some missing pieces that will be discussed in the workshop.

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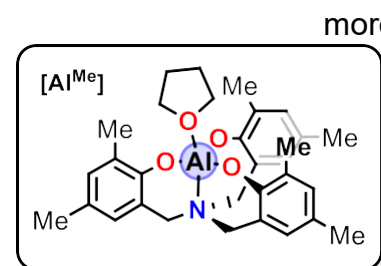
## Fatty Acid and Terpene Oxides as Versatile Precursors for Biobased Engineering Macromolecules

Francesco Della Monica,<sup>1</sup> Arianna Brandolese,<sup>1</sup> Ilaria Grimaldi,<sup>1</sup> Lorenz Dittrich,<sup>1</sup> Carles Bo,<sup>1</sup> Alba Villar-Yanez,<sup>1</sup> Fernando Bravo<sup>1</sup> and Arjan W. Kleij<sup>1,2</sup>

<sup>1</sup> Institute of Chemical Research of Catalonia (ICIQ-Cerca), Barcelona Institute of Science & Technology (BIST), Av. Països Catalans 16, 43007 – Tarragona (Spain). E-  
[akleij@iciq.es](mailto:akleij@iciq.es)

<sup>2</sup> Catalan Institute of Research and Advanced Studies (ICREA), Pg. Lluís Companys 23, 08010 – Barcelona (Spain)

In this contribution, we will demonstrate the key role of catalyst design to facilitate the challenging coupling of epoxy monomer derived from typically sterically demanding structures that involve fatty acid or terpene scaffolds. The developed catalytic processes allow for various novel macromolecular functionalized architectures with a primary focus on polyesters and polycarbonates. The pendent functionality will be demonstrated to be of use in advanced curing, modification and/or elaboration towards more advanced polymer backbones, thereby enabling the utilization as biomass as a starting point for more sustainable polymer and material development [1- 3]. Key examples of these achievements are illustrated, with a major focus on practical process conditions and catalyst systems that are easily accessed and have a modular character (see for an example the Figure included).



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A. Brandolese, F. Della Monica, M. À. Pericàs, A. W. Kleij, *Macromolecules* **2022**, 55, 2566–2573.

F. Della Monica, A. W. Kleij, *ACS Sustainable Chem. Engin.* **2021**, 9, 2619-2625.

## Terpene-Based Hybrid Polycarbonates via Co/Terpolymerization of Biobased Epoxides and CO<sub>2</sub>: Access to Cross-Linked Polycarbonates with Enhanced Thermal Stability and Functionality

Thirusangumurugan Senthamarai,<sup>1</sup> and Arjan W. Kleij<sup>1,2</sup>

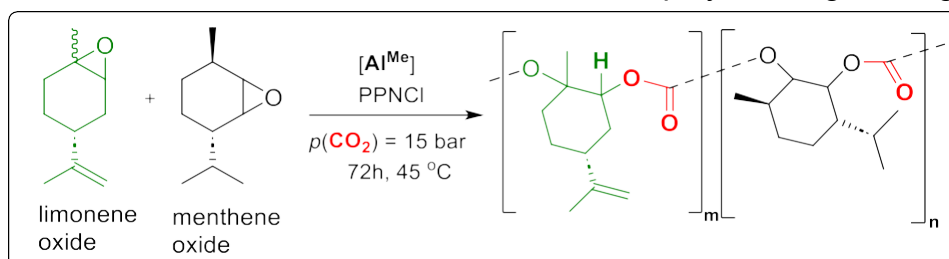
<sup>1</sup> Institute of Chemical Research of Catalonia (ICIQ), the Barcelona Institute of Science and Technology (BIST), Av. Països Catalans 16, 43007 - Tarragona, Spain.

<sup>2</sup> Catalan Institute of Research and Advanced Studies (ICREA), Pg. Lluís Companys 23, 08010 Barcelona, Spain

tsenthamarai@iciq.es

Biopolymers have received much attention due to their potential recyclability, non-toxicity, sustainability, and biodegradation potential. The utilization of biodegradable polymers and/or biobased polymers is a prominent approach to alleviate the problems associated to the use and disposal of synthetic plastics derived from fossil fuels.<sup>1</sup> The use of carbon dioxide (CO<sub>2</sub>) as a carbon feedstock in organic synthesis and polymer science, and the replacement of fossil fuel feedstock for natural, renewable ones in polymer engineering have become

important aspects of modern research.<sup>2</sup> We



report here

terpolymers derived from renewable terpenes (*D*-limonene and 2-menthene) producing polycarbonates with a controlled ratio between each monomer. As catalyst, a simple binary combination of an Al(III) aminotriphenolate complex and PPNCI was used to afford the targeted biobased polycarbonates with typically high carbonate content, good monomer conversion and a controllable amount of olefinic groups in the terpolymer backbone.<sup>3</sup>

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## Vegetable Oil-Based Hyperbranched Materials – Synthesis and Derivatization Towards New Properties

Guillaume Chollet,<sup>1</sup> Marie Reulier,<sup>1</sup> Etienne Grau,<sup>2</sup> Henri Cramail,<sup>2</sup> Boris Bizet<sup>1</sup>

<sup>1</sup> ITERG, 11 Rue Gaspard Monge – ZA PESSAC – CANEJAN, 33610 CANEJAN, France

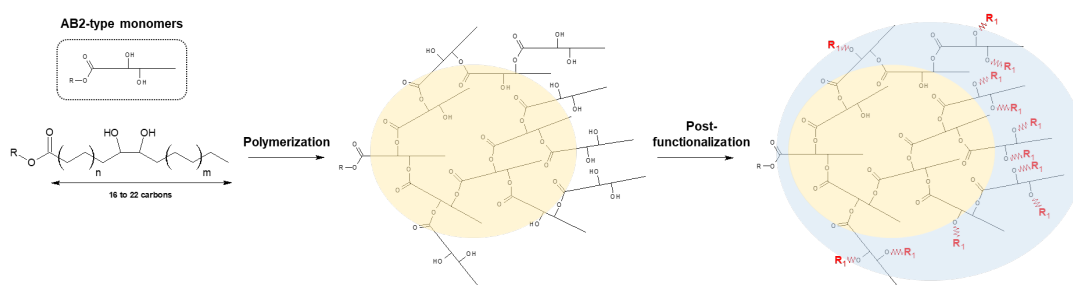
<sup>2</sup> LCPO – UMR 5629, Université de Bordeaux – CNRS – Bordeaux INP, 16 Avenue Pey Berland, 33607 PESSAC, France

b.bizet@iterg.com

Hyperbranched polymers are a very specific class of dendritic materials. Similar to dendrimers, they are composed of a highly branched polymer structure, with a high outer functionality. Having a compact and globular geometry, remarkable properties can be achieved among which a lower viscosity (compared to their linear homologues of lower molar masses), higher solubilities, and a much higher functionality.

For about a decade, ITERG – in partnership with LCPO – has fostered the development of a vegetable oil-based platform for the design of specific molecules that can be precursors of hyperbranched polymers. Starting from various fatty acid sources containing 16 to 22 carbons stemming from edible and non-edible resources, fully bio-based “AB2”-type monomers have thus been designed and synthesized at the kilolab and pilot scale, paving the way towards the one-step green synthesis of hyperbranched polyesters. Polymers of various microstructures could be obtained, also at the kilolab (and even pilot) scale for some structures.

Taking advantage of their high functionality, several derivatization pathways have been investigated to broaden the scope of applicability of the obtained polymers. Applications as additives, surfactants, plasticizers can be envisioned. ITERG is now looking for interested partners in order to better position this novel technology on an applicative range.



## New (Sustainable) Pathways to Integrate Sulfur Functional Groups into Renewable Based Polymers

Cuong-Minh-Quoc Le, Adam Woodhouse, Yagmur Deniz Karatas, Bercis, Pektas, Rayane Toutaoui, Gautier Schrodj, and Hatice Mutlu

Institut de Science des Matériaux de Mulhouse, UMR 7361 CNRS/Université de Haute Alsace, 15 rue Jean Starcky, Mulhouse France  
hatice.mutlu@uha.fr

In recent years, industrial chemical production of petroleum-based polymers is being re-imagined, and the principles of Green, Sustainable and Circular chemistries, respectively,<sup>1</sup> are leading the redesign of industrial-scale processes to minimize the environmental footprint of those polymers. In other words, the formidable quest for the development of straightforward methods to enable access to the targeted polymers from simple and readily available, potentially renewable-based, starting materials with higher efficiency and minimal waste is escalating. In parallel, there is urgent need to replace those non-degradable polymers with up-/re-cyclable materials.<sup>2</sup> Relevantly, the synthesis of sulfur-containing polymers<sup>3</sup> has sparked great interest in polymer science, in view of both novel structure construction and property exploration by exhibiting a wide range of attractive features, up-/re-cyclability being the most preferable (compared to their oxygen counter partners). Thus, in the light of the current need for more sustainable and affordable materials displaying thermo-mechanical properties on par with current mass-produced polymers, we have established practical methods by using readily available and inexpensive renewable monomers to deliver various S-containing polymers. Their potential as promising, bio-derived and up-/re-cyclable material platform will be emphasized by presenting exemplary applications.

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## **Optimisation of the RAFT Polymerisation of Pendant Fatty Acid Acrylamide Monomers Synthesised via the Base Catalysed Transesterification of Plant Oils**

Oliver Harris, Ryan Larder, Helen Willcock, Fiona Hatton

Loughborough University, Department of Materials, Loughborough, United Kingdom

[o.harris@lboro.ac.uk](mailto:o.harris@lboro.ac.uk)

Polymeric materials based on fatty acids have a combination of characteristics (functionality, hydrophobicity, tuneable  $T_g$ ) that give them great potential as partially renewable, high value added materials. Previous studies have demonstrated the synthesis and free radical polymerisation of a range of unsaturated pendant fatty acid acrylamide monomers via an industrially relevant transesterification reaction. However, despite its similarity to the well understood biodiesel reaction there are key differences that currently limit the yield and purity of products. Additionally, no research has yet been conducted on the use of these monomers in controlled radical polymerisation systems (e.g. RAFT) which would enable the design of high performance materials. In this work we investigated the base catalysed transesterification of four different plant oils with N-hydroxyethyl acrylamide by conducting kinetics experiments, investigating potential side reactions and by greatly improving characterisation and isolation of the target product. Impure brine washed monomers were intentionally used in the kinetics studies of RAFT polymerisations in order to evaluate the suitability of the existing monomer synthesis method as a practical and sustainable solution. Successful control of the reactions by RAFT was assessed by using  $^1\text{H}$  NMR spectroscopy and GPC to obtain monomer conversion,  $M_n$  and  $\bar{M}_w$  data. Further developments in this area will enable higher yield syntheses of monomers suitable for RAFT polymerisation, and the range of thermal properties ( $12 < T_g < 56$  °C and  $-16 < T_m < 47$ ) accessible dependent on the pendant fatty acid could open the doors for a wide range of unique, renewably sourced materials.

## Non-Isocyanate Polyurethanes From Terpene and Erythritol Feedstock Using Organocatalysis and Thiol-ene Reaction

F. Clara M. Scheelje<sup>1</sup>, Michael A. R. Meier<sup>1,2</sup>

<sup>1</sup> Laboratory of Applied Chemistry, Institute of Organic Chemistry (IOC), Karlsruhe Institute of Technology (KIT), Straße am Forum 7, 76131, Karlsruhe, Germany

<sup>2</sup> Laboratory of Applied Chemistry, Institute of Biological and Chemical Systems - Functional Molecular Systems (IBCS-FMS), Karlsruhe Institute of Technology (KIT), Hermann-von-Helmholtz-Platz 1, 76344, Eggenstein-Leopoldshafen, Germany  
clara.scheelje@kit.edu

A synthesis route towards building blocks for non-isocyanate polyurethanes (NIPUs) using renewable starting materials is presented. Based on terpenes and erythritol, five-membered cyclic carbonates were synthesized and ring-opened by amines with terminal double bonds. In this way, a scope of monomers bearing one or two urethane moieties and two terminal double bonds was obtained. In case of terpene-based cyclic carbonates, the ring-opening was promoted by thiourea compounds as benign and efficient organocatalysts. The obtained AA monomers served as building blocks for NIPU synthesis via step-growth thiol-ene polyaddition. The reaction with different dithiols yielded linear NIPUs under mild conditions. Variation of the dithiol and amine led to polymers with different properties, with  $M_n$  of up to  $31 \text{ kg} \cdot \text{mol}^{-1}$  and  $T_g$ 's ranging from 1 to 29 °C. Introduction of polyfunctional thiols enabled the implementation of these urethane building blocks into cross-linked NIPU materials.

## New Concepts for the Synthesis and Valorization of Fatty Acids

Lukas Gooßen

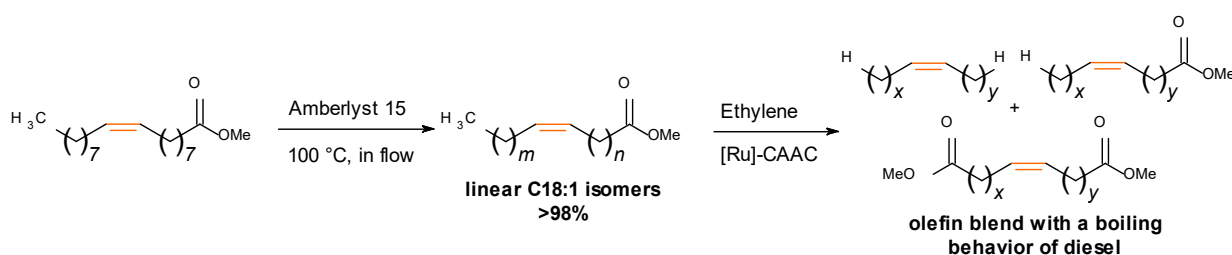
Ruhr-Universität Bochum, Fakultät für Chemie und Biochemie, Universitätsstr. 150, 44801

Bochum, Germany.

Lukas.Goossen@ruhr-uni-bochum.de

We have recently introduced isomerizing metathesis as a tool for the valorization of vegetable oils. Initially, a bimetallic palladium/ruthenium catalyst system was utilized to access industrially useful functionalized olefin blends with adjustable chain lengths from non-purified fatty acid esters.<sup>[1]</sup> The catalyst system allows to generate biofuel suitable for use in unmodified Diesel engines.<sup>[2]</sup> To further increase the economic feasibility of this process, we have now investigated a two-step sequential isomerization and ethenolysis process<sup>[3]</sup> (Figure 1). The double bond migration in methyl oleate could be efficiently performed in a flow reactor over solid catalysts.<sup>[4]</sup> Ethenolysis of the C18:1 isomers in the presence of 50ppm of CAAC-ligated Hoveyda-Grubbs catalyst furnishes an olefin blend that closely resembles diesel fuel in its boiling behavior.

We have also developed a decarboxylative ketonization process that allows coupling of short-chain carboxylic acids generated from CO<sub>2</sub> and water via a power-to-chemical process. The products have been converted further into fatty acid esters.<sup>[5,6]</sup>



**Figure 1.** Concept of the stepwise double bond isomerization-metathesis of methyl oleate.

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## Homogeneous Catalysis for the Functionalization of Oleochemicals and Their Derivatives into Value-Added Products

Sébastien Tilloy

Univ. Artois, CNRS, Centrale Lille, Univ. Lille, UMR 8181, Unité de Catalyse et Chimie du Solide (UCCS), rue Jean Souvraz, SP 18, 62300 Lens, France  
sebastien.tilloy@univ-artois.fr

As part of the transition to a post-petroleum chemistry, chemists are increasingly turning to the production of new molecules and innovative materials from bio-based resources such as vegetable oils.<sup>[1]</sup> Indeed, vegetable oils and their derivatives are particularly interesting building blocks due to the possibility of the carbon–carbon double bond modification. Among the various possibilities, such as hydrogenation, hydroformylation, hydroaminomethylation, metathesis, epoxidation... the hydrohydroxymethylation (HHM) reaction has been much less studied. HHM (also known as reductive hydroformylation) consists of the catalytic transformation of carbon–carbon double bonds into primary alcohol functions via a hydroformylation reaction followed by a hydrogenation reaction.<sup>[2]</sup>

Recent developments in this field will be presented, with a focus on some vegetable oils and their derivatives (castor, jojoba or linseed oil).<sup>[3,4]</sup> The possibility of recycling the catalytic system and the applications of the polyols produced will also be discussed.<sup>[5,6]</sup>

<sup>[1]</sup> U. Biermann, U. T. Bornscheuer, I. Feussner, M. A. R. Meier, J. O. Metzger, *Angew. Chem. Int. Ed.* 2021, 60, 20144 <sup>[2]</sup> G. M. Torres, R. Frauenlob, R. Franke and A. Börner, *Catal. Sci. Technol.*, 2015, 5, 34 <sup>[3]</sup> C. Becquet, F. Berche, H. Bricout, E. Monflier, S. Tilloy, *ACS Sustain Chem Eng*, 2021, 9, 9444 <sup>[4]</sup> C. Becquet, M. Ferreira, H. Bricout, B. Quienne, S. Caillol, E. Monflier and S. Tilloy *Green Chemistry*, 2022, 24, 7906 <sup>[5]</sup> A. El Mouat, C. Becquet, J. Ternel, M. Ferreira, H. Bricout, E. Monflier, M. Lahcini, and S. Tilloy *ACS Sustainable Chem. Eng*, 2022, 10, 34, 11310 <sup>[6]</sup> A. El Mouat, T. El Assimi, M. Raihane, J. Ternel, H. Bricout, E. Monflier, S. Tilloy, M. Lahcini, *Progress in Organic Coatings* 179 (2023) 107513

## Photoexcited Nitroarenes as Ozone Surrogates

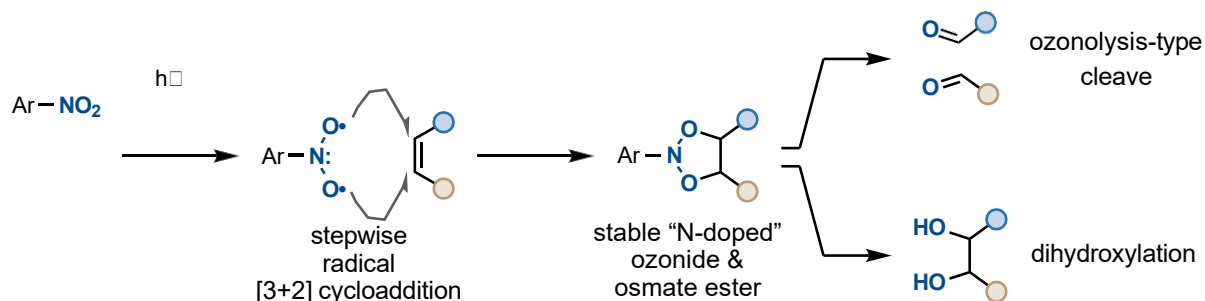
Prof Daniele Leonori

Institute of Organic Chemistry, RWTH Aachen University, Aachen, Germany

daniele.leonori@rtwh-aachen.de • @LeonoriLab • www.leonorigroup.com

Nitroaromatics are widely available feedstocks that are routinely used for the preparation of anilines. I will present our most recent work that demonstrates how these species can be used, upon blue light irradiation, to promote the ozonolysis-style cleavage<sup>[1]</sup> and dihydroxylation<sup>[2]</sup> of olefins.

The presentation will focus on the most important mechanistic aspects that control the reactivity of photoexcited nitroarenes and will also illustrate how their reactivity can be modulated to achieve selectivity for olefin functionalization in derivatives containing more than one olefin.



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## **Fats and Oils - An Important Part of Transforming Chemistry to a Circular Economy**

Manuel Häußler,<sup>1,2</sup> Nicolas Friese,<sup>1</sup> Melissa de Rossi,<sup>1</sup> Patrick Rathenow<sup>1</sup>

<sup>1</sup> Max-Planck-Institute of colloids and interfaces, Potsdam, Germany

<sup>2</sup> Center for the Transformation of Chemistry (CTC), Delitzsch, Germany

manuel.haeussler@mpikg.mpg.de

The chemical industry is on the verge of a major transformation. Driven by a growing global population and a corresponding surge in consumption, the demand for carbon-based chemistry is escalating. Today, the industry consumes more than 500 million metric tons of carbon per year, a figure that is expected to exceed 1 gigaton by 2050. This forecast presents a dual challenge: not only will we need to source more than twice as much carbon for chemical production as we do today, but it is imperative that we do so in a sustainable manner. The shift from a linear economy, reliant on fossil-based raw materials, to a genuinely circular economy is critical in this regard. In this talk, I will highlight the key role of fats and oils, both first generation sources and waste materials, in the transition of the chemical industry, with a special focus on circular plastics. In addition, the talk will introduce the newly established "Center for the Transformation of Chemistry", a major research institute currently being established in Delitzsch, Leuna and Merseburg to spearhead this transformative journey.



# **Abstracts**

## **Part 2: Posters**

## Continuous Selective FAME Hydrogenation Utilizing a Microcapillary Reactor

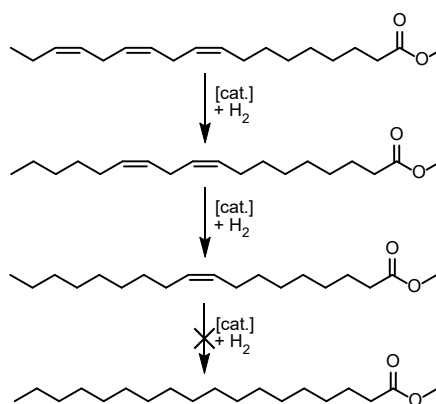
Florian Lehmann<sup>1</sup>, Niclas von Vietinghoff<sup>1</sup>, Takenobu Nakai<sup>1</sup>, Lennard Nebel<sup>1</sup>, Peter Pey<sup>1</sup>,  
Karl Steffen Wulle<sup>1</sup>, Dieter Vogt<sup>1</sup>, Thomas Seidensticker<sup>1</sup>

<sup>1</sup> Laboratory of Industrial Chemistry, Department of Biochemical and Chemical Engineering, TU Dortmund University, Dortmund, Germany

florian.lehmann@tu-dortmund.de

Selective hydrogenation of polyunsaturated fatty acid derivatives is a crucial step prior to carrying out further oleochemic reactions like hydroformylation or metathesis to produce biobased value added products with increased activity and selectivity<sup>[1]</sup>. With this step, the crude raw material consisting of different substances with varying degree of saturation can be harmonised to mostly monounsaturated compounds.

Combining selective homogeneous catalysts with reaction engineering extends the potential of the applied reaction systems. For instance, microcapillary reactors offer several advantages compared to conventional stirred tank reactors: Especially high surface-to-volume ratios enhance mass and energy transport<sup>[2]</sup>. They also allow for continuous small scale production. The feasibility of this reactor concept for the homogeneously catalyzed selective hydrogenation of fatty acid methyl esters (FAME, scheme 1) is evaluated, especially in terms of mass transport limitations, feeding of reaction gas and in comparison to a standard batch reaction.



**Scheme 1: Simplified intended reaction scheme.**

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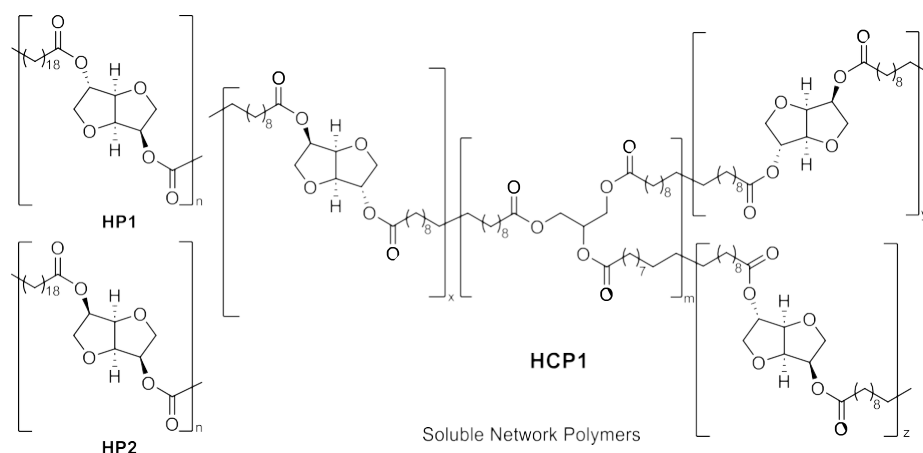
## Biobased Linear/Network Aliphatic Polyesters Exhibiting Promising Tensile Properties

Kotohiro Nomura,<sup>1,\*</sup> Kosaku Tao,<sup>2</sup> Mika Kojima,<sup>1</sup> Xiuxiu Wang,<sup>1</sup> Lance O.P. Go,<sup>1</sup> Daisuke Shimoyama,<sup>1</sup> Mohamed M. Abdellatif,<sup>1</sup> Seiji Higashi,<sup>2</sup> and Hiroshi Hirano<sup>2</sup>

<sup>1</sup>Tokyo Metropolitan University, Hachioji, Tokyo, Japan;

<sup>2</sup>Osaka Research Institute of Industrial Science and Technology (ORIST), Osaka, Japan  
ktnomura@tmu.ac.jp

Aliphatic polyesters derived from plant resources attract considerable attention<sup>1-3</sup> in terms of circular economy due to rather facile ability of chemical recycling (through transesterification etc.)<sup>4</sup> than conventional polyolefins. We herein report synthesis of high molecular weight polyesters prepared by acyclic diene metathesis (ADMET) polymerization of bis(undec-10-enoate) with isosorbide, isomannide, and the subsequent hydrogenation.<sup>4-6</sup> The tensile strength and the strain (elongation at break) in the resultant polymers (HP1, HP2) increased upon increasing the molar mass,<sup>6</sup> and the strain further increased when the polymerizations were conducted in the presence of glycerol based cross-linker (HCP1).<sup>7</sup> Details will be introduced in the symposium.



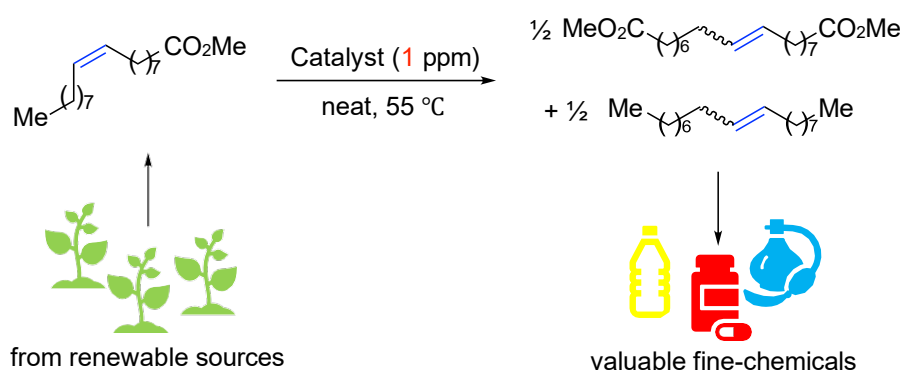
- [1] (a) Nomura, K.; Awang, N. W. B. *ACS Sustainable Chem. Eng.* **2021**, *9*, 5486. (b) Nomura, K. *Catalysts* **2024**, *14*, 97.  
 [2] Stempfle, F.; Ortmann, P.; Mecking, S. *Chem. Rev.* **2016**, *116*, 4597.  
 [3] ADMET polymerization: Fokou, P. A.; Meier, M. A. R. *J. Am. Chem. Soc.* **2009**, *131*, 1664.  
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 [5] Nomura, K. et al. *ACS Omega* **2020**, *5*, 18301; **2023**, *8*, 7222; **2024**, *9*, 9109.  
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## New Cyclic(Alkyl)(Amino)Carbene-Based Olefin Metathesis Catalysts for Green Chemical Conversion of Fatty Acids Derivatives

Filip Struzik

Biological and Chemical Research Centre, Faculty of Chemistry, University of Warsaw,  
 Żwirki i Wigury Street 101, 02–089 Warsaw, Poland  
 f.struzik@student.uw.edu.pl

Plants (i.e. soy, alga, canola) are excellent renewable source of fatty acids, which can be easily converted into widely used methyl ester analogues (FAMEs). On the other hand, olefin metathesis is powerful method to form new carbon–carbon double bonds with great group tolerance. That make this reaction useful to obtain valuable for industry substrates from FAMEs. Key to implement this transformation on wide scale is usage of stable catalyst with low loading. During research in Grela's Group, we were able to synthesise new Ru-based catalyst with cyclic (alkyl)(amino)carbene ligands—air and moisture stable complexes, which can be use in ppm loading level.<sup>1,2</sup>



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<sup>2</sup> Karol Grela et al., Novel Ruthenium Complexes, Method of Their Synthesis, Intermediate Compounds Used in This Method, Method of Their Synthesis and the Use of Novel Ruthenium Complexes in Olefin Metathesis Reactions, WO2023248205A1, filed 23 June 2023, and issued 28 December 2023.



## Conversion of Plant Oil Feedstocks via Continuous Flow Ethenolysis and Double Bond Isomerization

Lara N. Holderied, Inigo Göttker-Schnetmann and Stefan Mecking

Department of Chemistry, Chair of Chemical Material Science, University of Konstanz,  
78464 Konstanz, Germany  
Lara.holderied@uni-konstanz.de

1-Olefins are key intermediates of the chemical industry. They are produced today from fossil feedstocks nearly exclusively. Alternative sustainable feedstocks are desirable.<sup>1</sup> A combination of chain cleavage by ethenolysis and of double bond isomerization generates 1-olefins from plant oil feedstocks.<sup>2,3</sup> To obtain only desired terminal unsaturated products, these steps need to be carried out in a sequential fashion. Solid rather than soluble catalysts are useful to enable a sequential catalysis continuous flow scheme. This contribution reports solid-supported molecular catalysts for the catalytic upgrading of plant oil feedstocks *via* olefin metathesis.

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- (2) Ohlmann D. M., Tschauder N., Stockis J.-P., Goossen K., Dierker M., Goossen L.J. Isomerizing olefin metathesis as a strategy to access defined distributions of unsaturated compounds from fatty acids *J. Am. Chem. Soc.* **2012**, *134*, 13716-73729.
- (3) De Roo S., Einsiedler F., Mecking S. Catalytic Biorefining of Natural Oils to Basic Olefinic Building Blocks of Proven Chemical Valorization Schemes *Angew. Chem. Int. Ed.* **2023**, *62*, e202219222.

## Synthesis of Biobased Poly(Ester Amide)s From Long-Chain Fatty Acids and Oligopeptides

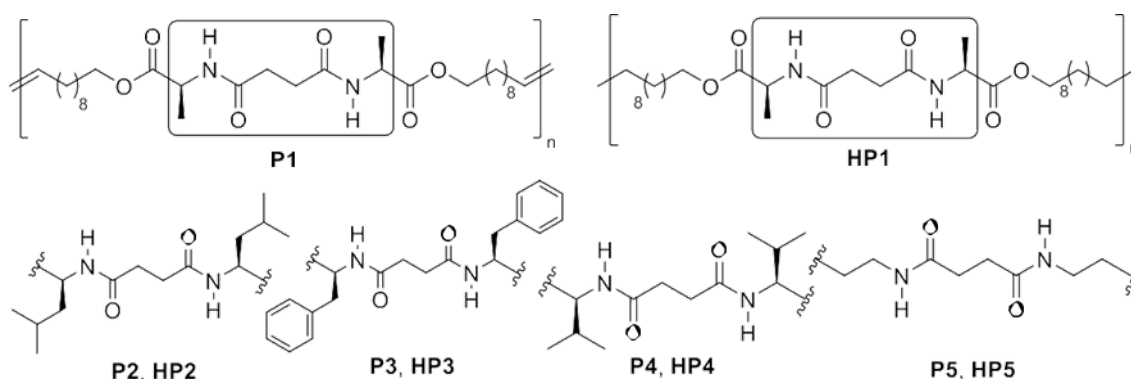
Gaku Miyamoto, Mohamed Mehawed Abdellatif, and Kotohiro Nomura\*

Department of Chemistry, Tokyo Metropolitan University, Hachioji, Tokyo, Japan

miyamoto-gaku@ed.tmu.ac.jp

Research on recyclable, high-performance biobased long chain aliphatic polyesters, polyamides, derived from inedible vegetable oils, has attracted much attention in the field of polymer chemistry and materials chemistry.<sup>1,2</sup> We herein report synthesis of the biobased poly(ester amide)s by acyclic diene metathesis (ADMET) polymerization and the subsequent hydrogenation, since physical properties in these polymers are influenced by the interpolymer communications.

These poly(ester amide)s (**HP1-5**, **P1-5**, chart shown below) were prepared by ADMET polymerization<sup>3-5</sup> of symmetric nonconjugated dienes, consisting of fatty acid esters (castor oil) and a series of oligopeptides through a ester linkage, using ruthenium-carbene catalyst and the subsequent tandem hydrogenation. It tuned out that thermal and mechanical properties of the resultant polymers are strongly affected by the ester amide segment employed. Details will be introduced in the symposium.



[1] (a) Nomura, K.; Awang, N. W. B. *ACS Sustainable Chem. Eng.* **2021**, *9*, 5486-5505. (b) Nomura, K. *Catalysts* **2024**, *14*, 97.

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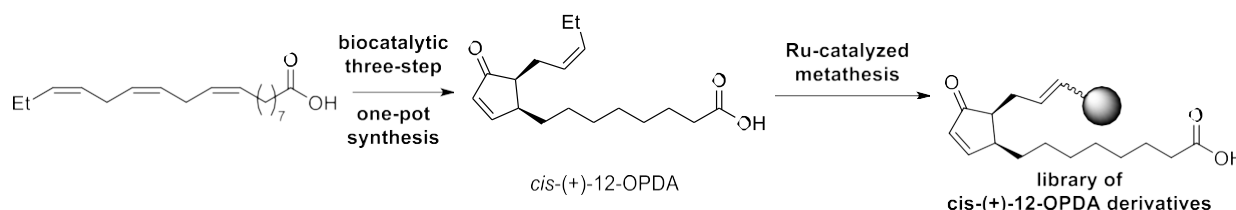
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## Biocatalytic Synthesis of *cis*-(+)-12-Oxophytodienoic Acid and Derivatization Using Metathesis

Maike Bittmann, Tim Lukas Guntelmann, Madita Knieper, Jana Löwe, Andrea Viehhauser, Harald Gröger, Karl-Josef Dietz

Bielefeld University, Bielefeld, Germany

The plant metabolite *cis*-(+)-12-oxophytodienoic acid (*cis*-(+)-12-OPDA) serves as a precursor in the biosynthesis of jasmonic acid, but also raised itself interest as a plant hormon due to its biological activity in plants. Recently we developed a biocatalytic organic synthesis of *cis*-(+)-12-OPDA based on the three biosynthetic steps starting from linolenic acid. Within a one-pot process in the presence of a lipoxygenase, allene oxide synthase and allene oxide cyclase, the desired *cis*-(+)-12-OPDA was formed with >99% conversion, at least 90% selectivity and ≤10% of side products in combination with a high diastereoselectivity with the diastereomeric ratio being at least 90:10.<sup>[1]</sup>



**Scheme 1.** Chemoenzymatic synthesis of *cis*-(+)-12-OPDA derivatives

In this contribution we report on process development of this route as well as on the utilization of the synthesized *cis*-(+)-12-OPDA for preparing derivatives with modified side-chains at the 2-position by means of a ruthenium-catalyzed cross-alkene metathesis reaction as a chemocatalytic key step (Scheme 1).<sup>[1]</sup> These reactions proceed with conversions of up to >99% and enable a synthetic access to a compound library of new *cis*-(+)-12-OPDA derivatives with different substitution pattern at this side-chain. Thus, such a combination of a biocatalytic synthesis of *cis*-(+)-12-OPDA and subsequent metal-catalyzed derivatization gives access to a structurally unique oxylipin library, which has been utilized for biological structure-activity relationship studies.

References:

[1] J. Löwe, K.-J. Dietz, H. Gröger, *Adv. Science* **2020**, 7, 1902973.

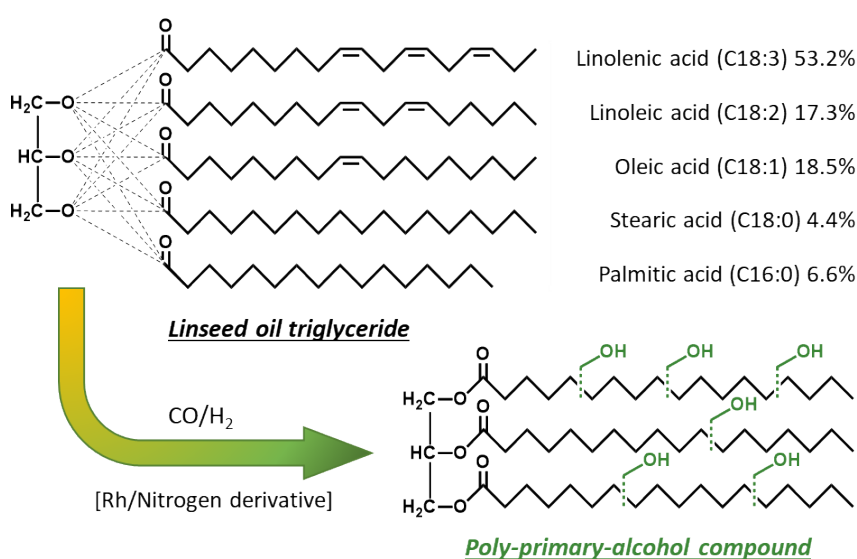
## Production of Polyols From Linseed Oil by Reductive Hydroformylation

Walid Abdallah, Michel Ferreira, Hervé Bricout, Eric Monflier, Sébastien Tilloy

Univ. Artois, CNRS, Centrale Lille, Univ. Lille, UMR 8181, Unité de Catalyse et Chimie du Solide (UCCS), rue Jean Souvraz, SP 18, 62300 Lens, France

walid.abdallah@univ-artois.fr

Vegetable oils are renewable resources that can replace fossil fuels in the production of chemical compounds. Among the various oils available on the market, linseed oil is full of interest. Indeed, this triglyceride is predominantly composed of five fatty acids (palmitic acid, stearic acid, oleic acid, linoleic acid, and linolenic acid, as illustrated in Scheme 1 [1]. Our objective is to catalytically valorize linseed oil and its derivatives (mainly the methyl esters of linoleic and linolenic acids) by hydrohydroxymethylation which consists of the conversion of carbon-carbon double bonds into primary alcohols via a tandem hydroformylation/hydrogenation sequence. The catalytic system is based on rhodium associated to nitrogen derivatives. Our goal is to optimize the catalytic system (nature of nitrogen derivatives) and operational conditions (temperature, pressure) to obtain the most efficient system. The compounds obtained through this process could serve as valuable monomers for synthesizing polyesters or polyurethanes.



*Scheme 1 : HHM of Linseed oil.*

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## Poly(Menthene Carbonate): Synthesis and Depolymerization Studies

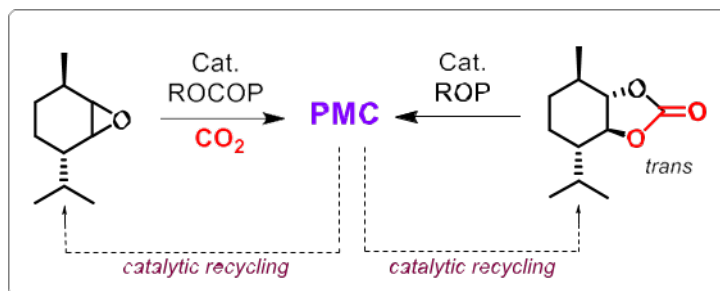
Enrico Lanaro<sup>1</sup> and Arjan W. Kleij<sup>1,2</sup>

<sup>1</sup> Institute of Chemical Research of Catalonia (ICIQ), the Barcelona Institute of Science & Technology (BIST), Av. Països Catalans 16, 43007 - Tarragona, Spain.

E-mail: elanaro@iciq.es

<sup>2</sup> Catalan Institute of Research and Advanced Studies (ICREA), Pg. Lluís Companys 23, 08010 Barcelona, Spain

Most plastics are not recycled, causing not only problems and pollution to our eco-systems, but also resulting into a large waste of resources.<sup>1</sup> This project involves the valorization of biocarbon feedstock through catalytic processes, and specifically the conversion of menthol into poly(menthene carbonate), PMC. Two different and possible polymerization pathways are studied: the ring-opening copolymerization (ROCOP) of 2-menthene oxide and CO<sub>2</sub> catalyzed by aminotriphenolate Al/Fe complexes,<sup>2</sup> and the organocatalytic ROP of *trans*-menthene carbonate (partially) obtained from a biomass source. Subsequently, the depolymerization and subsequent repolymerization of PMC is studied, taking advantage of recent progress attained in the field.<sup>3</sup> Our long-term project aims are to develop a circular use of novel biobased polycarbonates (such as PMC) and having a detailed mechanistic understanding of the polymer build-up and break-down processes.



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## Catalyst-Free Synthesis of Bio-Derived Vitrimers Decorated with Dynamic Thioacetal Linkages

Yagmur Deniz Karatas,<sup>1</sup> Cuong-Minh-Quoc Le,<sup>1</sup> Gautier Schrodj,<sup>1</sup> and Hatice Mutlu<sup>1</sup>

<sup>1</sup> Institut de Science des Matériaux de Mulhouse, UMR 7361 CNRS/Université de Haute Alsace, 15 rue Jean Starcky, Mulhouse France; hatice.mutlu@uha.fr

According to processing property, conventional polymer materials are classified into thermoplastics and thermosets. On the one hand, thermoplastics can be recycled multiple times without losing their properties, which makes them more environmentally friendly, on the other hand, thermosets are non-processable and non-recyclable but possess prominent mechanical properties. Vitrimers, introduced by Leibler et al. in 2011, were developed with the idea of bridging the difference between thermoplastics and thermosets. [1,2]. In this regard, many different chemistries (ranging from furan-maleimide chemistry to allyl sulfide exchange reactions) have been applied or purposely developed for vitrimer synthesis. As part of our group's interest towards vitrimer synthesis in addition to the exploration and development of new reversible chemistry platforms for their design, we were inspired by the light-induced thiol-aldehyde condensation reaction to deliver, in a straightforward way, highly dynamic thioacetal linkages, starting from very simple (potentially bio-derived) precursors (**Figure 1**). Thus, series of different aldehyde derivatives (such as vanillin, syringaldehyde, 4-hydroxybenzaldehyde, and cinnamaldehyde) have been systematically probed for their ability to produce dynamic thioacetal linkages on a multi-gram scale

(up to ~100 g) in high yield (96 %). The obtained polymeric material demonstrated tuneable viscoelastic behaviour and thermal properties. Moreover, the vitrimers displayed reprocessability over

minimum of 2 destruction/healing cycles. Our findings indicate that vitrimers can be easily constructed from diverse bio-derived aldehyde derivatives in the presence of multifunctional thiols. Moreover, their tunable properties and high-performance thermomechanical behaviours make them appealing for applications in the aerospace, automotive, electronics and biomedical sectors.

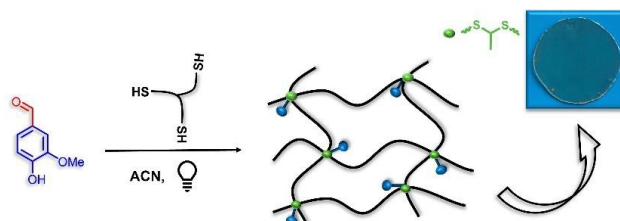


Fig. 1: The representative catalyst-free, light-induced reaction using bio-derived aldehydes and thiols for the design of vitrimers decorated with dynamic thioacetal linkages

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## **Project SymbioLoop: How to Convert Waste Fats and Oils into Fully Circular Plastics**

Nico Friese<sup>1</sup>, Melissa De Rossi<sup>1</sup>, Patrick Rathenow<sup>1</sup>, Manuel Häußler\*<sup>1,2</sup>

<sup>1</sup> Max-Planck-Institute of colloids and interfaces, Potsdam, Germany

<sup>2</sup> Center for the Transformation of Chemistry (CTC), Delitzsch, Germany

nico.friese@mpikg.mpg.de

The SymbioLoop consortium of the SPRIN-D Challenge "Circular Biomanufacturing" focuses on the conversion of local waste fats and oils, such as used cooking oil or larvae fat, into sustainable, high-performance plastics. We present the upstreaming of industrial wastes into chemical intermediates that can be converted into various polycondensates capable of replacing fossil-based products for various applications. A unique feature of our materials is their mild chemical recycling through hydrolysis to the parent building blocks. This not only allows the recovery of valuable monomers, but also enables new product designs where composites, e.g. with natural fibers, would be fully recyclable.

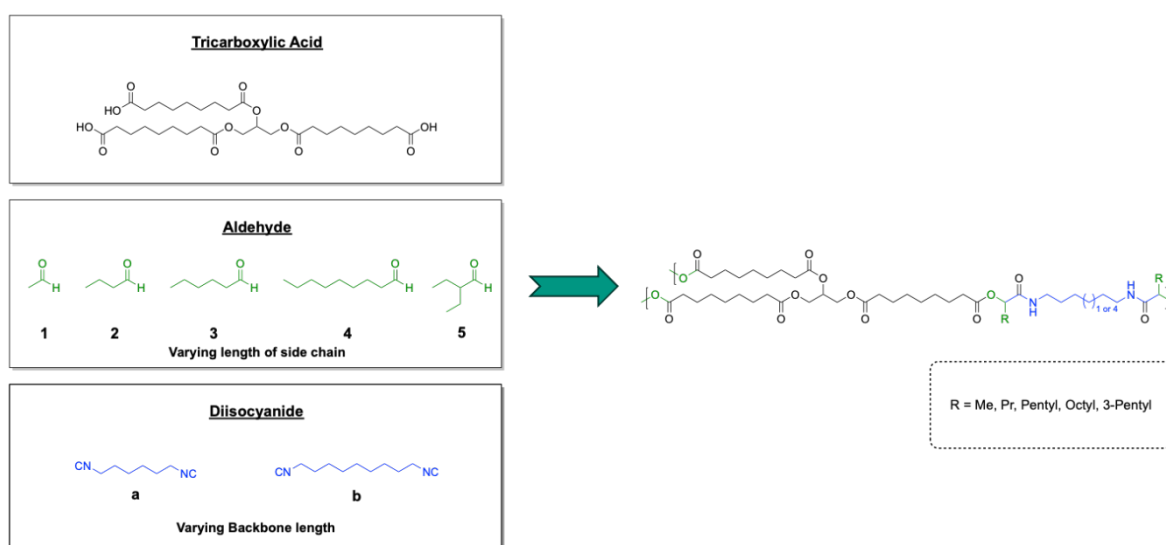
## Sunflower Oil Based Thermosets via the Passerini Three Component Reaction

Luis Santos Correa, Michael A. R. Meier

Institute of Biological and Chemical Systems – Functional Molecular Systems (IBCS-FMS),  
Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

luis.correa@kit.edu

Tricarboxylic acids are molecules of interest for the synthesis of highly cross-linked polymers. Herein, a synthesis route to a novel triacid from high oleic sunflower oil is described by applying a ruthenium catalyzed oxidative cleavage of its double bonds. This bio-based triacid was used for the synthesis of cross-linked materials *via* the Passerini three component reaction. Ten different polymeric materials were synthesized by variation of aldehyde and diisocyanide components and subsequently characterized *via* IR spectroscopy, swelling tests, thermogravimetric analysis, differential scanning calorimetry, and tensile tests. The characterization of the insoluble polymeric networks was complemented by the synthesis of model compounds to enable analysis in solution *via* nuclear magnetic resonance spectroscopy. Due to the fast curing of all polymers at room temperature, adhesive tests were performed to demonstrate their potential application as glues.





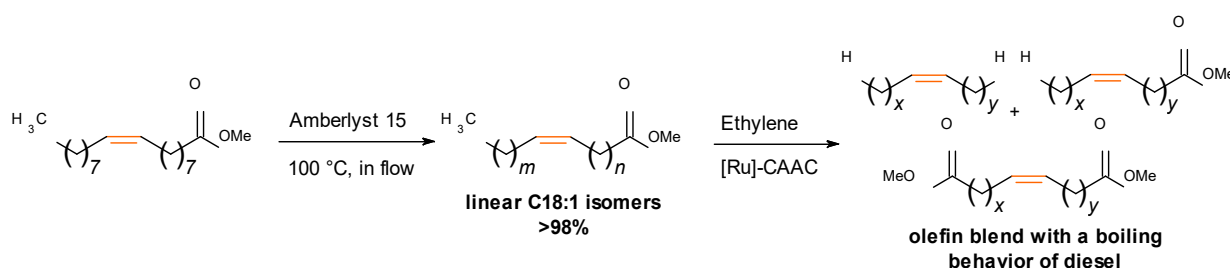
## Diesel Fuel From Methyl Oleate by Sequential Double Bond Isomerization and Metathesis

Mykhailo Kondratiuk, Lukas Gooßen

Ruhr-Universität Bochum, Fakultät für Chemie und Biochemie, Universitätsstr. 150, 44801 Bochum, Germany.

mykhailo.kondratiuk@rub.de, lukas.goossen@rub.de

Within recent years, we have been investigating isomerizing metathesis reactions as tools for the valorization of vegetable oils as renewable resources.<sup>[1]</sup> A bimetallic palladium<sup>[2]</sup>/ruthenium catalyst system enables the synthesis of industrially useful functionalized olefin blends with tailored chain lengths. In this context, technical quality fatty acids can be employed in isomerizing metathesis processes,<sup>[3]</sup> allowing for the production of a biofuel identical to diesel under mild conditions.<sup>[4]</sup> To further increase the economic feasibility of this process, we have investigated the use of a two-step sequential isomerization and ethenolysis upon mono-unsaturated fatty acid scaffolds<sup>[5]</sup> (Figure 1). The double bond migration in methyl oleate could be efficiently performed in a flow reactor over zeolites<sup>[6]</sup> or ion-exchange resins. With subsequent ethenolysis of the C18:1 isomers over a CAAC-ligated Hoveyda-Grubbs catalyst at ppm levels, we could again generate an olefin blend that closely resembles diesel fuel in its boiling behavior.



**Figure 1.** Concept of the stepwise double bond isomerization-metathesis of methyl oleate.

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## Novel Highly Efficient Catalysts for Ethenolysis

Rafał Gawin<sup>1</sup>, Andrzej Tracz<sup>1</sup>, Patryk Krajczyk<sup>1</sup>, Anna Kozakiewicz-Piekarz<sup>2</sup>, Juan Pablo Martínez<sup>3</sup>, Bartosz Trzaskowski<sup>3</sup>

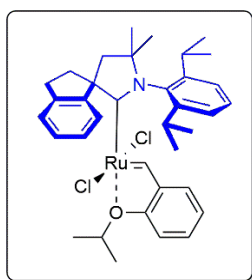
<sup>1</sup> Apeiron Synthesis SA, Duńska 9, 54-427 Wrocław, Poland

<sup>2</sup> Faculty of Chemistry, Nicolaus Copernicus University in Toruń, Gagarina 7, 87-100 Toruń, Poland

<sup>3</sup> Centre of New Technologies, University of Warsaw, Banacha 2c, 02-097 Warszawa, Poland

rafal.gawin@apeiron-synthesis.com

Ruthenium-based Hoveyda-type olefin metathesis catalysts bearing novel rigid spiro-Cyclic Alkyl Amino Carbenes (CAACs) have been developed. They are characterized by exceptional stability towards decomposition through  $\beta$ -elimination and bimolecular pathways, thus enabling unprecedented efficiency in cross-metathesis of seed oil-derived fatty acid esters with ethylene (ethenolysis). Catalyst loading as low as 100 ppb was applied to the ethenolysis of the model substrate methyl oleate, leading to a turnover number (TON) of 2.6 million. Ethenolysis of methyl esters derived from high oleic sunflower oil and rapeseed oil – readily available on an industrial scale, inexpensive and renewable feedstocks – was for the first time effectively carried out with 0.5 ppm catalyst loading with TON as high as 964 000.

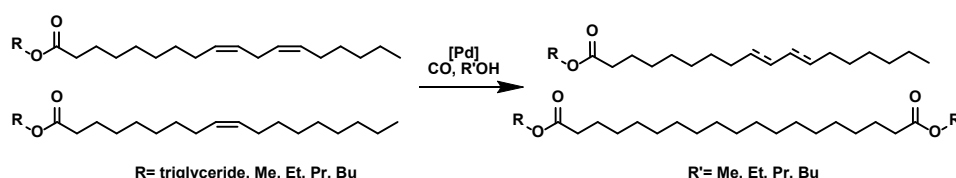


# On the Degree of Unsaturation in the Pd-Catalyzed Alkoxy carbonylation of Oleochemicals – New Approaches and Insights

Hannes Wegener, Florian Lehmann, Thomas Seidensticker

Laboratory of Industrial Chemistry, Department of Biochemical and Chemical Engineering,  
TU Dortmund University, Dortmund, Germany  
hannes.wegener@tu-dortmund.de

Polyunsaturated fatty acids (PUFAs), especially linoleic acid, are one of the major components, along with oleic acid, of readily available vegetable oils such as rapeseed and soybean oil. While oleic acid has been extensively studied, the functionalization of PUFAs remains a significant challenge. Despite notable advancements, there are still difficulties in the conversion of PUFAs, as evidenced by the complex nature of reactions such as the alkoxycarbonylation (Scheme 1). As the literature sheds light on the existing hurdles, it becomes increasingly clear that novel approaches and deeper insights are essential to achieve efficient conversion and overcome the inhibiting effects associated with PUFAs.<sup>[1]</sup> In this context, our research addresses new perspectives on isomerization and innovative strategies for a better conversion.



**Scheme 2: Isomerizing alkoxy carbonylation and isomerization of unsaturated fatty acids with various alcohols and palladium as a homogeneous catalyst.**

This work includes novel approaches, including one-pot substrate pretreatment and innovative separation techniques. In addition, we will study the intricacies of isomerization and intermediate formation. The isolation and characterization of such intermediates is one part, while their utilization and transformation will also be investigated. Through these efforts, we aim to advance the use of renewable resources in the chemical industry.

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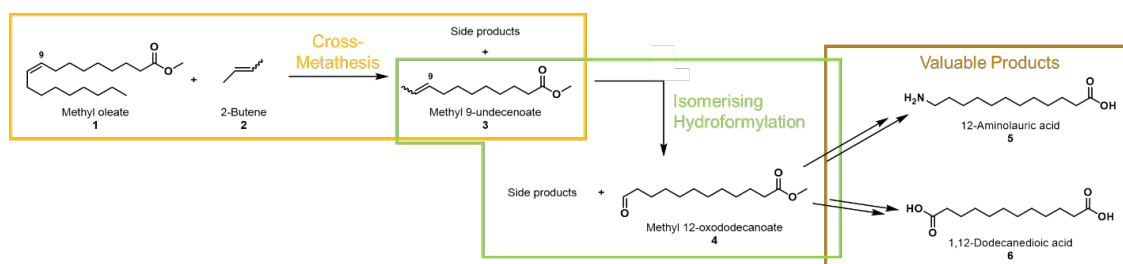
## C<sub>12</sub>-Polymer Precursors From Local Plant Oils Through Cross Metathesis and Isomerizing Hydroformylation

Jerome Hommes, Dieter Vogt, Thomas Seidensticker

Laboratory of Industrial Chemistry, Department of Biochemical and Chemical Engineering, TU Dortmund University, Dortmund, Germany

jerome.hommes@tu-dortmund.de

Dodecanedioic acid (**6**) and C<sub>12</sub> polycondensates such as polyamide 12 are versatile and valuable materials, but are currently produced based on fossil raw materials. Methyl 12-oxododecanoate (**4**) is a potential platform chemical for the production of C<sub>12</sub> polycondensates based on renewable resources, as it can be converted into a wide range of molecules, like aminolauric acid (**5**) or **6**.<sup>[1]</sup> Until now, **4** has been produced from methyl 10-undecenoate, which is made from ricinoleic acid,<sup>[2]</sup> available from castor oil. However, castor oil has to be imported and is more expensive than local plant oils such as rapeseed, sunflower, etc. Therefore, we propose an alternative route to **4** (Scheme 1) based on methyl oleate (**1**), available from local plant oils, via cross metathesis and isomerising hydroformylation.



Scheme 1: Reaction network for the synthesis of methyl 12-oxododecanoate (**4**) by cross-metathesis of methyl oleate (**1**) to methyl 9-undecenoate (**3**) and isomerising hydroformylation, as well as subsequent products aminolauric acid (**5**) and Dodecanedioic acid (**6**).

Methyl 9-undecenoate (**3**) is first produced from methyl oleate (**1**) by cross-metathesis with 2-butene (**2**), which is contained in crude butane obtained during the purification of 1-butene. The product has no terminal double bond, which is why an isomerising hydroformylation is carried out to obtain the linear bifunctional product (**4**). Rhodium or cobalt are to be used as catalyst metals with bidentate phosphorus ligands, which can enable subsequent catalyst recycling.

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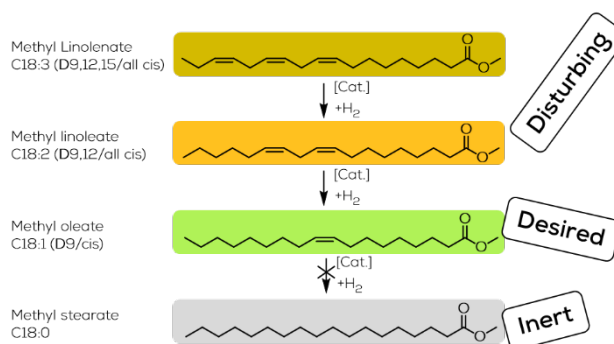
## From Fundamental Research to Start-up Founding



Maximilian Spiekermann, Max Krause, Jens Ehlhardt, Marén Schwandt, Thilo Wattrodt,  
Thomas Seidensticker

Simplyfined, Laboratory of Industrial Chemistry, Department of Biochemical and Chemical  
Engineering, TU Dortmund University, Dortmund, Germany

The ever-present global shortage of fossil resources increases the demand for sustainable alternatives. Not only as renewable feedstock for fuels but also as a starting material for chemical conversions leading to plastics and other functionalized compounds.<sup>[1]</sup> High potential for functionalization of C-C double bonds in oleochemicals by various homogeneous transition metal catalysts has been shown in several cases in the past.<sup>[1,2]</sup> Partial hydrogenation of poly-unsaturated fatty acid derivatives will be a key step for the utilization of various renewable resources as replacement for petrochemical intermediates.<sup>[3]</sup> Increasing the degree of saturation to mono-unsaturated fatty acid derivatives paves the way for various chemical reactions like epoxidation, hydroformylation or ethenolysis to shorter terminal alkenes or other functionalized intermediates. Simplyfined (founding in process) is a start-up company originating from TU Dortmund University. Their process utilizes a novel catalytic system. The catalyst already shows high activity at temperatures up to 50°C and high selectivity to the monounsaturated fatty compound in the mixture. Optimization of reaction conditions led to reaction times below 3 minutes. Process simulation and modeling lead to resilient prediction of product composition and highly competitive product cost.



**Figure 1: Stepwise partial hydrogenation of poly-unsaturated fatty acid methyl esters.**

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## Bio-Polyols for High-Performance Composites From Local Vegetable Oils

Johannes Stiehm, Anika Hegemann, Harald Gröger

Bielefeld University, Bielefeld, Germany

johannes\_raphael.stiehm@uni-bielefeld.de

Vegetable oils represent as an attractive biorenewable feedstock for preparing multifunctionalized compounds that are of interest for the formation of polymers such as polyesters and polyurethanes. High-oleic sunflower oil is an interesting building block because of its low price and readily availability. Furthermore, it contains oleic acid as desired unsaturated fatty acid for functionalization at a very high content. From a molecular point of view, the introduction of two or three hydroxy groups per vegetable oil molecule is crucial for obtaining monomers for the synthesis of polyesters and polyurethanes. A straightforward approach for these vegetable oil-derived monomers is based on chemocatalytic multiple hydroxy-functionalization by means of initial hydroformylation and subsequent hydrogenation of the formed aldehyde moieties. As for the hydroformylation key step, this technology has already been widely applied for plant oils.[1] For hydroformylation processes, in general rhodium catalysts turned out to be highly efficient.[2] A current challenge is the development of heterogeneous rhodium catalysts, which enable efficient separation of the catalytic component from the reaction mixture after the hydroformylation. In this contribution, we present a comparison of methods for the heterogenization of rhodium catalysts for hydroformylation exemplified for hydroformylation of high-oleic acid sunflower oil. Furthermore, optimization tests with the two-phase reaction systems for this hydroformylation reaction are presented.

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## **Interreg VI A-Project 'Biotech Talent Unlocked' (31042)**

Mark Rüschen gen. Klaas, Bernd Schmietenknop

University of Applied Sciences Emden/Leer, Emden, Germany

bernd.schmietenknop@hs-emden-leer.de

### **Aim of the Project:**

The Ems-Dollart-Region distinguishes as region with a strong background in agriculture. Lots of biomass is available and the expertise in logistics and processing is on a high level. To reach the goal of the EU Green Deal it is important to reduce Glas-House-Gas emissions (GHG). GHG emissions of agricultural processing can be reduced using the following ways:

- Needed energy should be sustainable (biogas, wind, solar, hydrogen)
- Fermentation or bioconversion of agricultural products and even waste to valorize biomass to high-end markets
- Replacement of petrochemical products by agricultural products

One of the most important ingredients for this program is human capital. A crossborder educational bio-network has been founded to make the Ems-Dollart-Region more visible and more attractive for students or young employees.

In three workpackages, a network is under construction to organize student's workshops in bio-economy and excursions to companies of the Ems-Dollart-Region. Than joint student-projects and the opening of selected modules for students of the partners are the next steps. Themes can be for example biodiesel-production or bioconversion of renewable materials. The final aim is to design joint educational activities. These will be intra- and extracurricular and open for bachelor- and master students.

Participating partners are the Hanze University of Applied Science, NHL Stenden Hogeschool, 3N Kompetenzzentrum e.V, Hochschule Emden/Leer, Rijksuniversiteit Groningen and BVIO B.V. (BBENG). Leadpartner is BIO Cooperative B.V.

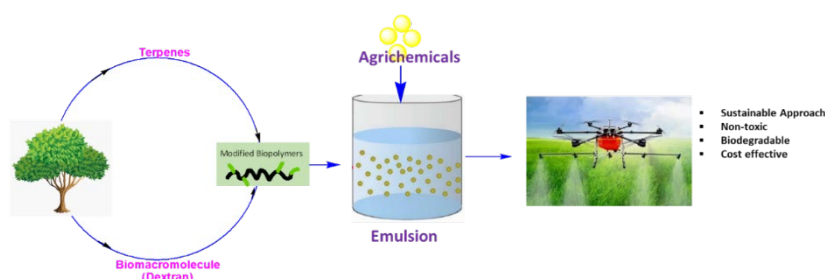
## Design and Development of Biodegradable Macromolecule Self-Assembly for Agriculture Formulations

Vinay Chauhan, Pankaj Sharma, Isha Soni

School of Advanced Chemical Sciences, Shoolini University, Solan 173229, India

Email: chauhanvinay1985@gmail.com

In the realm of agricultural advancements, the utilization of polysaccharides, such as dextran, has emerged as a pivotal innovation for developing environmentally friendly polymers [1]. Here, we have developed the environmentally friendly polymers suitable for agricultural formulations by combining hydrophilic dextran with hydrophobic terpenes. The process involved the coupling of amino-functionalized dextran (Dextran-NH<sub>2</sub>) with carboxyl-functionalized monoterpenes (both cyclic and acyclic) using Novozymes chemistry at ambient temperature. The desired structures were characterized using <sup>1</sup>H and DOSY NMR spectroscopy. The synthesized polymer-based system was employed as an emulsifier for herbicide formulations in aqueous solutions at varying weight percentages. Emulsion stability was assessed through visual observation, and droplet size was determined using optical microscopy, with droplet sizes ranging from 5-30 µm. The emulsions remained stable for up to 28 days. Additionally, emulsions formed with Dextran-based polymers coupled with cyclic terpene derivatives exhibited superior stability compared to those formulated with acyclic terpenes. Hence, such type of new bio-based emulsifier system underscores the potential for sustainable and efficient agricultural applications.



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## **Synthesis and Characterization of PBT-Based Thermoplastic Copolyesters Containing a Renewable Fatty-Acid-Derived Soft Block**

Apostolos A. Karanastasis<sup>1</sup>, Victoria Safin<sup>1</sup>, Subin Damodaran<sup>2</sup>, and Louis M. Pitet<sup>1\*</sup>

<sup>1</sup>Advanced Functional Polymers (AFP) Laboratory, Institute for Materials Research (IMO), Hasselt University, Martelarenlaan 42, 3500 Hasselt, Belgium

<sup>2</sup>Tosoh Bioscience, GmbH, Im Leuschnerpark 4, 64347 Griesheim, Germany  
subin.damodaran@tosoh.com

Thermoplastic copolyesters (TPCs) serve as pivotal constituents in numerous high-performance applications characterized by stringent requirements for thermal stability and mechanical robustness. Segmented multiblock architectures, amalgamating semicrystalline poly(butylene terephthalate) (PBT) segments with various low glass transition temperature (T<sub>g</sub>) soft blocks, are frequently employed for such demanding applications. Conventionally, these segmented copolymers are synthesized from pristine feedstocks predominantly derived from fossil-fuel sources. This study introduces a facile, one-pot synthetic strategy for the fabrication of TPCs. High-molar mass poly(ethylene terephthalate) recyclate (rPET) is employed in conjunction with a hydrophobic fatty acid dimer diol flexible segment. Transesterification serves as the pivotal mechanism for generating a multiblock architecture. The multiblock polymer systems were then characterized with Size exclusion chromatography coupled with refractive index and light scattering detection. Due to the different conformation of the polymers with that of the polystyrene standards in solution, the molar mass and the dispersity from the light scattering measurements were in-line with expectations based on the step-growth mechanism in comparison to the results from refractive index detection.

## **Synthesis of Biobased and Biodegradable Polyurethane Coating from Olive Pomace Oil for Controlled-Release Fertilizers**

Gökhan Çaylı<sup>1</sup>, Fatih Özönder<sup>2</sup>

<sup>1</sup> Istanbul University-Cerrahpaşa, Faculty of Engineering, Engineering Sciences, Istanbul, Turkey

<sup>2</sup> Istanbul Technical University, Department of Polymer Science and Technology, Istanbul, Turkey

Email address of presenting author: ozonder21@itu.edu.tr

Chemical fertilizers, particularly urea with its high nitrogen content, are essential in modern agriculture. However, their rapid solubilization poses environmental risks such as groundwater contamination and atmospheric pollution. In response to these challenges, a biobased coating material has been developed. The solid leftover material after pressing olives to release their oil is called olive pomace. Olive pomace may contain at least 75% solid material that is composed of 50% cellulose, 10% protein, and 15 % crude oil. The composition of the crude pomace oil is similar to olive oil. In this study, pomace oil is reacted with diethanolamine to synthesize pomace oil diethanolamide. That compound is used for the synthesis of biobased polyurethanes. Hexamethylene diisocyanate and methylene diphenyl diisocyanate are used for polyurethane synthesis. Biobased polymers are very valuable coating materials for controlled-release fertilizers. Urea was used as a model compound. Urea beads were coated using rotary drum mixers. The thickness of the polyurethane layer is measured as 30 µm. By using biodegradable polyurethane coating, the release of the urea beads can be extended to 6 months.

**Keywords:** Urea, Olive pomace oil, Diethanolamide, biobased polyurethane, Controlled-release fertilizers, Coating material

# List of participants



# List of participants

## **Walid Abdallah**

University of Artois, UCCS  
Faculté des Sciences  
rue Jean Souvraz  
LENS 62307  
France  
walid.abdallah@univ-artois.fr

## **Cengiz Azap**

Apeiron Synthesis S. A.  
ul. Duńska 9  
54-427 Wrocław  
Poland  
cengiz.azap@apeiron-synthesis.com

## **Dr. Ursula Biermann**

abiosus e.V.  
Philipp Scheidemann Str. 21  
26133 Oldenburg  
Germany  
Ursula.biermann@uni-oldenburg.de

## **Maike Bittmann**

Bielefeld University  
Faculty of Chemistry, Chair of Industrial  
Organic Chemistry and Biotechnology  
Universitätsstraße 25  
33615 Bielefeld  
Germany  
maike.bittmann@uni-bielefeld.de

## **Boris B. Bizet**

ITERG  
Chimie du Végétal et physico-chimie  
11 rue Gaspard Monge - ZA Pessac  
Canejan  
33610 CANEJAN  
France  
iterg@iterg.com

## **Dr. Clemens Blasius**

KLK Kolb  
R&D  
Langestraat 167  
7491 AE Delden  
Netherlands  
Clemens.blasius@kolb.ch

## **Prof. Dr. Uwe Bornscheuer**

University of Greifswald, Institute of  
Biochemistry  
Dept. of Biotechnology & Enzyme  
Catalysis  
Felix-Hausdorff-Str. 4  
17489 Greifswald  
Germany  
uwe.bornscheuer@uni-greifswald.de

## **Dr. Gökhan Cayli**

Istanbul University-Cerrahpasa  
Department of Engineering Sciences  
İstanbul Üniversitesi-Cerrahpaşa  
Mühendislik Fakültesi Üniversite  
Mahallesi Bağlariçi Caddesi No:7  
34320 Istanbul  
Turkey  
gokhan.cayli@iuc.edu.tr

## **Dr. Vinay Chauhan**

Shoolini University  
School of Advanced Chemical Sciences  
Solan Solan  
India  
chauhanvinay1985@gmail.com

## **Dr. Subin Damadaran**

Tosoh Bioscience GmbH  
Im Leuschnerpark 4  
64347 Griesheim  
Germany  
Subin.damodaran@tosoh.com

## **Melissa De Rossi**

Max Planck Institute of colloids and  
interfaces  
Biomolecular System  
Am Mühlenberg 1, 14476 Potsdam  
10247 Berlin  
Germany  
Melissa.DeRossi@mpikg.mpg.de

**Prof. Martino Di Serio**

Università di Napoli Federico II  
Chemical Sciences  
via Cintia  
80126 Napoli  
Italy  
diserio@unina.it

**Dr. Markus Dierker**

BASF  
Henkelstrasse 67  
Dusseldorf 40551  
Germany  
markus.dierker@basf.com

**Dr. Angelino Doppiu**

Umicore AG & Co. KG  
Precious Metals Chemistry  
Rodenbacher Chaussee 4  
Hanau-Wolf 63457  
Germany  
angelino.doppiu@eu.umicore.com

**Nico Friese**

Max-Planck-institute  
Colloids and Interfaces  
Am Mühlenberg, 1  
14476 Potsdam  
Germany  
nico.friese@mpikg.mpg.de

**Franziska Fuhl**

TU Dortmund  
BCI/TC  
Emil-Figge 66  
44227 Dortmund  
Germany  
franziska.fuhl@tu-dortmund.de

**Dr. Rafal Gawin**

Apeiron Synthesis S.A.  
Duńska 9  
Wrocław 54-427  
Poland  
rafal.gawin@apeiron-synthesis.com

**Dr. Torsten Germer**

Robert Kraemer GmbH  
Zum Roten Han 9  
26180 Rastede  
Germany  
torsten.germer@rokra.com

**Prof. Dr. Lukas J Goossen**

Ruhr-Universität Bochum  
Fakultät für Chemie und Biochemie  
Universitätsstraße 150  
44801 Bochum  
Germany  
lukas.goossen@rub.de

**Prof. Dr. Andreas Greiner**

Universität Bayreuth  
Lehrstuhl für Makromolekulare Chemie  
Universitätsstrasse 30  
95440 Bayreuth  
Germany  
greiner@uni-bayreuth.de

**Prof. Dr. Karol Grela**

University of Warsaw  
Faculty of Chemistry / Biological and  
Chemical Research Centre  
Żwirki i Wigury 101  
02-089 Warsaw  
Poland  
klgre@uw.edu.pl

**Prof. Dr. Harald Gröger**

Bielefeld University  
Faculty of Chemistry, Chair of Industrial  
Organic Chemistry and Biotechnology  
Universitätsstr. 25  
33615 Bielefeld  
Germany  
harald.groeger@uni-bielefeld.de

**Dr. Harald Häger**

Evonik Oxeno GmbH & Co. KG  
Corporate Service  
Postfach 10 14 40 Gebäudenummer:  
1033  
45744 Marl  
Germany  
haeger@semigator.de

**Oliver J. Harris**

Loughborough University  
Department of Materials  
Epinal Way  
LE11 3TU Loughborough  
United Kingdom  
o.harris@gmail.com

**Dr. Manuel Häußler**

Max-Planck-Institute of colloids and  
interfaces & CTC  
Biomolecular Systems  
Am Mühlenberg 1  
14476 Potsdam  
Germany  
manuel.haeussler@mpikg.mpg.de

**Anika Hegemann**

University Bielefeld  
Faculty of chemistry  
Universitätsstr. 25  
33615 Bielefeld  
Germany  
anika.hegemann@uni-bielefeld.de

**Dr. Prisca Henheik**

Wiley-VCH  
Boschstr. 12  
69469 Weinheim  
Germany  
phenheik@wiley.com

**Lara Holderied**

University of Konstanz  
Department of Chemistry  
Universitätsstrasse 10  
78464 Konstanz  
Germany  
lara.holderied@uni-konstanz.de

**Prof. Dr. Frank Hollmann**

Delft University of Technology  
Department of Biotechnology  
van der Maasweg 9  
2629HZ Delft  
Netherlands  
f.hollmann@tudelft.nl

**Jerome Hommes**

TU Dortmund, BCI  
Lehrstuhl für Technische Chemie  
Emil-Figge-Str. 66  
44227 Dortmund  
jerome.hommes@tu-dortmund.de

**Yagmur Deniz Karatas**

Université de Haute-Alsace  
Institut de Science des Matériaux de  
Mulhouse  
15 rue Jean Starcky  
68057 Mulhouse  
France  
yagmur-deniz.karatas@uha.fr

**Heinrich Katz**

Hermetia Baruth GmbH  
An der Birkenpfuhlheide 10  
15837 Baruth  
Germany  
h.katz@hermetia.de

**Dr. Oscar Kelly**

BYK  
Flat A Crosbie House  
Frodsham WA67HE  
United Kingdom  
oscar.kelly@altana.com

**Prof. Dr. Arjan Willem Kleij**

ICIQ Tarragona  
Research group Kleij  
Av. Paisos Catalans 16  
Spain  
akleij@iciq.es

**Dr. Azra Kocaarslan**

Karlsruhe Institute of Technology  
Institute of Chemical Technology and  
Polymer Chemistry  
Engesserstraße 15  
Karlsruhe  
Germany  
azra.kocaarslan@kit.edu

**Dr. Andreas Kohl**

Verbio SE  
Ritterstrasse 23  
04109 Leipzig  
Germany  
andreas.kohl@verbio.de

**Mykhailo Kondratiuk**  
Ruhr-Universität Bochum  
Organische Chemie I  
Universitätsstr. 150  
44801 Bochum  
Germany  
mykhailo.kondratiuk@rub.de

**Dr. Philipp Kratzer**  
Clariant Plastics & Coatings  
(Deutschland) GmbH  
86368 Gersthofen  
Germany  
philipp.kratzer@clariant.com

**Max Krause**  
TU Dortmund  
BCI  
Steinerstr. 10  
53225 Bonn  
Germany  
max.krause@simplyfined.com

**Enrico Lanaro**  
Institute of Chemical Research of  
Catalonia (ICIQ)  
Chemistry  
Avinguda dels Països Catalans, 16  
43007 Tarragona  
Spain  
elanaro@iciq.es

**Florian Lehmann**  
TU Dortmund University  
Laboratory of Industrial Chemistry  
Emil-Figge-Str. 66  
44227 Dortmund  
Germany  
florian.lehmann@tu-dortmund.de

**Prof. Dr. Daniele Leonori**  
Institute of Organic Chemistry  
RWTH Aachen University  
Landoltweg, 1  
52056 Aachen  
Germany  
daniele.leonori@rwth-aachen.de

**Dr. Jan Lorenzen**  
Technical University of Munich  
Werner Siemens-Chair of Synthetic  
Biotechnology  
Lichtenberg Str. 4  
85748 Garching bei München  
Germany  
jan.lorenzen@tum.de

**Dr. Ling Ma**  
Klüber Lubrication München SE & Co. KG  
Research and Development  
Geisenhausenerstraße 7  
81379 Munich  
Germany  
Ling.Ma@klueber.com

**Prof. Dr. Michael A. R. Meier**  
Karlsruhe Institute of Technology (KIT)  
Institute of Organic Chemistry  
Straße am Forum 7  
76131 Karlsruhe  
Germany  
m.a.r.meier@kit.edu

**Prof. Dr. Jürgen O. Metzger**  
abiosus e.V.  
Bloherfelder Str.239  
26129 Oldenburg  
Germany  
juergen.metzger@uni-oldenburg.de

**Gaku Miyamoto**  
Tokyo Metropolitan University  
Chemistry  
1-1, Minami-Ozawa, Hachioji-shi, Tokyo  
192 0397  
Japan  
miyamoto-gaku@ed.tmu.ac.jp

**Prof. Dr. Eric Monflier**  
Univ. Artois  
Faculté des Sciences Jean Perrin  
Rue Jean Souvraz  
62307 Lens  
France  
eric.monflier@univ-artois.fr



**Dr. Hatice Mutlu**

Université de Haute-Alsace  
Institut de Science des Matériaux de  
Mulhouse  
15, rue Jean Starcky  
68057 Mulhouse  
France  
hatice.mutlu@uha.fr

**Prof. Dr. Kotohiro Nomura**

Tokyo Metropolitan University  
Department of Chemistry  
1920397 Hachioji, Tokyo  
Japan  
ktnomura@tmu.ac.jp

**Fatih Özönder**

Istanbul Technical University  
Polymer Science and Technology  
Maslak  
Istanbul 34467  
Turkey  
ozonder94@gmail.com

**Dominik Pietschmann**

TU Dortmund  
BCI / TC  
Emil-Figge-Str., 66  
44227 Dortmund  
Germany  
dominik.pietschmann@tu-dortmund.de

**Poorva Ramadas**

Clariant Produkte (Deutschland) GmbH  
Industrieparkstr. 1  
84508 Burgkirchen  
Germany  
poorva.ramadas@clariant.com

**Patrick Rathenow**

Max Planck Institute of Colloids and  
Interfaces  
Biomaterial Systems  
Am Mühlberg 1  
14476 Potsdam-Golm  
Germany

**Thomas F. H. Roth**

TU Dortmund  
Technische Chemie  
Emil-Figge-Straße 66  
44127 Dortmund  
Germany  
thomas2.roth@tu-dortmund.de

**Luis Santos Correa**

Karlsruher Institut für Technologie (KIT)  
Institut für Organische Chemie (IOC)  
Fritz-Haber-Weg 6  
76131 Karlsruhe  
Germany  
luis.correa@kit.edu

**Naima Saou**

HOBUM Oleochemicals  
Research and development  
Konsul-Ritter-Straße 10  
21079 Hamburg  
Germany  
nsaou@hobum.de

**Clara Scheelje**

Karlsruhe Institute of Technology  
Institute of Organic Chemistry  
Straße am Forum 7  
76131 Karlsruhe  
Germany  
clara.scheelje@kit.edu

**Dr. Bernd Schmietenknop**

University of Applied Sciences  
Faculty of Technology  
Constantiaplatz 4  
26723 Emden  
Germany  
bernd.schmietenknop@hs-emden-leer.de

**Prof. Dr. Ulrich Schörken**

TH Köln  
Campus Leverkusen  
Campusplatz 1  
51379 Leverkusen  
Germany  
ulrich.schoerken@th-koeln.de

**Dr. Thomas Seidensticker**

Technische Universität Dortmund  
Laboratory of Industrial Chemistry  
Emil-Figge-Straße 66  
44227 Dortmund  
Germany  
thomas.seidensticker@tu-dortmund.de

**Dr. Thirusangu Murugan Senthamarai Ganapathy**

ICIQ  
Chemistry (Polymer)  
Avinguda dels Països Catalans, 16  
Tarragona 43007  
Spain  
tsenthamarai@iciq.es

**Maximilian L. Spiekermann**

Laboratory of Industrial Chemistry  
Chemical- and Biochemicalengineering,  
TU Dortmund University  
Emil-Figge-Straße 66  
44227 Dortmund  
Germany  
maximilian.spiekermann@tu-dortmund.de

**Johannes Stiehm**

University of Bielefeld  
Chemistry  
Universitätsstrasse 25  
33615 Bielefeld  
Germany  
johannes\_raphael.stiehm@uni-bielefeld.de

**Filip Struzik**

University of Warsaw  
Biological and Chemical Research  
Centre, Faculty of Chemistry  
Żwirki i Wigury Street 101  
02-089 Warsaw  
Poland  
f.struzik@student.uw.edu.pl

**Dr. Viktoriya Sturm**

Thünen Institute  
Institute of Market Analysis  
38116 Braunschweig  
Germany  
viktoriya.sturm@thuenen.de

**Dr. Kosaku Tao**

Osaka Research Institute of Industrial  
Science and Technology  
Polymer Processing Engineering Lab.,  
Research Division of Materials Sci. and  
Eng., Morinomiya Center  
1-6-50, Morinomiya, Joto-Ku  
5368553 Osaka-city, Osaka, Japan  
tao@orist.jp

**Prof. Dr. Sébastien Tilloy**

University of Artois, UCCS  
Faculté des Sciences  
rue Jean Souvraz  
62307 Lens  
France  
sebastien.tilloy@univ-artois.fr

**Peter Tollington**

Cargill BV  
Cargill Bio-Industrials Europe  
Evert van de Beekstraat 378  
1118CZ Schiphol  
Netherlands  
peter\_tollington@cargill.com

**Prof. Dr. Dieter Vogt**

TU Dortmund University  
Bio- and Chemical Engineering, Industrial  
Chemistry  
Emil-Figge-Strasse 66  
44227 Dortmund  
Germany  
dieter.vogt@tu-dortmund.de

**Hannes W. Wegener**

Laboratory of Industrial Chemistry  
Chemical- and Biochemicalengineering,  
TU Dortmund University  
Emil-Figge-Straße 66  
44227 Dortmund  
Germany  
hannes.wegener@tu-dortmund.de

**Dr. Philipp Weyrauch**

Insempra GmbH  
Am Klopferspitz 19a  
82152 Planegg-Martinsried  
Germany  
philip.veyrauch@insempra.bio

**Dr. Sophie C.C. Wiedemann**

Cargill Bio-Industrial BV

Buurtje 1

2802BE Gouda

Netherlands

sophie\_wiedemann@cargill.com

**Krzysztof Wilczyński**

Grupa Wilmar Marek Wilczyński S.K.A

Aleja Niepodległości 2E

55-020 Żórawina

Poland

k.wilczynski@wilmar-oils.pl