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ALFRED WERNER FUND, MASTER'S STUDENT SCHOLARSHIPS

The Alfred Werner Fund of the SCS Foundation was established in 2014 and continues the initiatives and projects of the former foundation 'Stiftung für Stipendien auf dem Gebiete der Chemie', also known as the 'Werner Stiftung'. The SCS Foundation is very proud to provide this program in collaboration with the Swiss chemical and pharmaceutical industry.
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Alfred Werner Master's Student Scholarships

The program invites scholarship applications to carry out Master degree studies in Chemistry or Biochemistry at a Swiss University or Federal Institute of Technology.

The Foundation offers up to ten scholarships of CHF 25'000 per year as a one-time contribution to the cost of the Master study program. This opportunity targets students from foreign countries in the top 10% of their undergraduate programs. The goal of the program is to bring in young talent to Swiss Universities or FIT or to keep them after the BSc studies in Switzerland.

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Alfred Werner Scholars 2018–2020

The committee of the Werner Fund is proud to announce the winners of the next term that starts in fall 2017:

- ***Michelle A. Gaspard***
  BSc at Polytechnique Montréal, Canada
  Support for her MSc studies at EPFL Lausanne
- ***Oleh Hordiichuk***
  BSc at Ivan Franko Nat. University of Lviv, Ukraine
  Support for his MSc studies at ETH Zürich
- ***Olivera Zivojinovic***
  BSc at University of Belgrade, Serbia
  Support for her MSc studies at ETH Zürich
- ***Dorina Morina***
  BSc at Kharkiv National University, Ukraine
  Support for her MSc studies at ETH Zürich
- ***Artem Kononenko***
  BSc at Institute of Organic Chemistry and Biochemistry, Prague, Czech Republic
  Support for his MSc studies at University of Geneva
- ***Jane Marsden***
  BSc at University of Limerick, Ireland
  Support for her MSc studies at University of Geneva
- ***Jeremy Wong***
  BSc at University of Toronto, Canada
  Support for his MSc studies at University of Geneva

Summaries of the Master Thesis from Students of the Term 2016–18

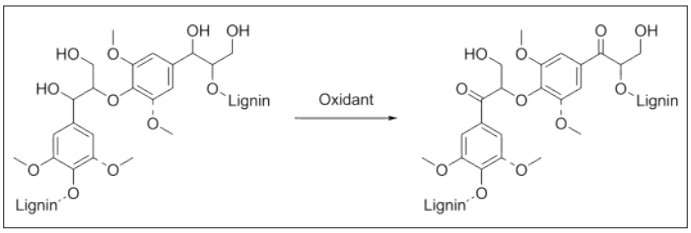
Jean Behaghel de Bueren
Nationality: *Belgian*
Bachelor at: *UCL*
Master at: *EPFL*
Master thesis supervisor:
Jeremy S. Luterbacher, Wu Lan

Biomass Conversion to Low Molecular Weight Aromatics *via* Selective Oxidation and Depolymerization of Protected Lignin

Lignin is the second most abundant biopolymer on earth after cellulose. Despite its abundance and promising characteristics as a feedstock for low molecular weight aromatic chemicals, it has not been used on an industrial scale. This stems from the challenge of extracting lignin from lignocellulosic biomass without degradation and from the difficulty to obtain materials from selective de-polymerization. On the one hand, selective oxidation of lignin has recently been reported as a promising pathway for the production of monomeric compounds.[1] On the other hand, a new ground-breaking method that uses protecting groups allows to extract uncondensed lignin with a biomass delignification close to 100%.[2] However, protection structures present on the lignin make it difficult to oxidize using current

processes. During this project, we developed a new oxidation method that allows protected lignin oxidation with high yields.

By deprotecting and oxidizing lignin in a one pot process, lignin condensation was avoided, and a high level of oxidation was reached. The α-OH of the β-aryl ether linkage in lignin was selectively oxidized with 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (DDQ) from a secondary alcohol to a ketone (see Fig. below).



In order to reduce the DDQ consumption, a catalytic system was designed. DDQ was regenerated *in situ* from the hydroquinone thanks to nitric acid as co-catalyst with molecular oxygen as the final oxidant.

After oxidation, lignin was depolymerized in aqueous formic acid under mild conditions (T = 110 °C) to form aromatic monomeric compounds with yields close to the theoretical one. Thanks to the combination of an effective lignin extraction and an efficient oxidation-depolymerization process, we achieved overall yields from lignin in biomass to low-molecular-weight compounds with yields 2.2–2.8 (catalytic and stoichiometric oxidation) times higher than previously reported ones.

[1] A. Rahimi, A. Ulbrich, J. J. Coon, S. S. Stahl, *Nature* **2014**, *515*, 249.
[2] L. Shuai, M. T. Amiri, Y. M. Questell-Santiago, F. Heroguel, Y. Li, H. Kim, R. Meilan, C. Chapple, J. Ralph, J. S. Luterbacher, *Science* **2016**, *354*, 329.

Future plans

After completion of my master thesis, I will join the EPFL startup 'Bloom Biorenewables' whose focus is on lignin valorization with the goal to transfer this new technology to industrial scale. My research will be on testing exotic feedstocks to find unique lignin-derived molecular mixtures. After that, I am considering pursuing a PhD in chemical engineering.

Dora Harangozo
Nationality: *Croatian*
Bachelor at: *J. J. Strossmayer University of Osijek*
Master at: *ETH Zurich*
Master thesis supervisor:
Dr. Bartosz Marcin Lewandowski
Wennemers group

Templated Length-controlled Oligomerization

In an attempt to achieve selective, length-controlled oligomerization reactions, a system based on rigid short peptides as templates was designed. Five peptides, each bearing a recognition site and activation sites for appropriately-functionalised molecular entities, were synthesised. Bifunctional small molecule substrates were prepared and their interactions with the peptidic templates were evaluated. Both covalent as well as non-covalent binding of the substrates to the peptides was successfully achieved. Furthermore it was demonstrated that substrates bound to the recognition sites on the peptides can be simultaneously activated for reactions with external agents.

DNA and RNA transcription and translation of the RNA code into the sequence of amino acids are essential for sustaining life. Molecular recognition and templating are key processes that enable the production of natural oligomers from the respective monomeric building blocks with a high degree of length and sequence control.[1] Yet despite tremendous progress in the fields of supramolecular chemistry and nanotechnology,[2] efficient length-controlled preparation of short oligomers using synthetic systems has not been accomplished to date.[3] This master thesis was aimed at utilizing molecular templates, based on rigid peptides to selectively recognize and activate small molecule substrates in order to prepare short oligomers with a high degree of substrate specificity and length control. My goal was to synthesize peptidic templates of different lengths and several small molecule substrates. The next step involved investigating interactions between the templates and the substrates (both dynamic covalent, as well as weak non-covalent interactions were probed). Upon developing an optimal set of conditions allowing to bind the substrates to the templates and activate them for reactions with one another, length-controlled oligomerization was attempted.

[1] S. Sainsbury, C. Bernecky, P. Cramer, *Nat. Rev. Mol. Cell Biol.* **2015**, *16*, 129.
[2] E. R. Kay, D. A. Leigh, F. Zerbetto, *Angew. Chem., Int. Ed.* **2007**, *46*, 72.
[3] K. S. Feldman, Y. B. J. Lee, *J. Am. Chem. Soc.* **1987**, *109*, 5850.

Future plans

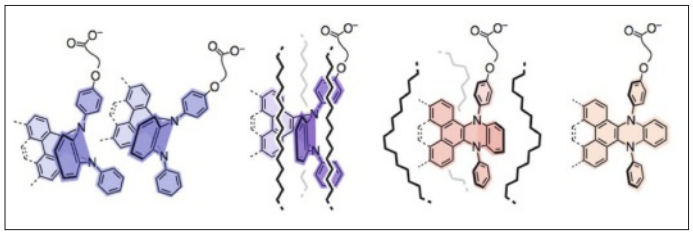
After completion of my MSc studies, I intend to do an internship in chemical industry. I would like to broaden my horizon and also to gain insight into industrial R&D. Later, I plan on applying for a PhD position at the Laboratory for Organic Chemistry or at the Institute for Pharmaceutical Sciences of ETH Zürich.

Heorhii Humeniuk
Nationality: *Ukraine*
Bachelor at: *University of Kyiv, Ukraine*
Master at: *University of Geneva*
Master thesis supervisor:
Prof. Stefan Matile

Dual-Emission Papillon Mechanophores for Sensing the Order of Lipid Bilayer Membranes

Papillon-shaped phenazines are introduced as novel mechanosensitive membrane probes. These compounds allow to discriminate micellar and membrane mediums with steady-state fluorescence spectroscopy, ratiometric two-photon excitation fluorescence microscopy (TPEFM) and bare eye. TPEFM is shown to be applicable for imaging of lipid domains in giant unilamellar vesicles (GUVs).

Fluorescence is the main tool for elucidating biophysical and biochemical processes occurring in cell membranes. Among all environment-sensitive probes,[1] there are only few examples of mechanophores[2a] responding to changes of forces, particularly tension in cells. They have been referred as 'flipper probes' to highlight the importance of a large molecular surface area to assure significant mechanosensitivity. The changes in fluorescence lifetime upon ground state untwisting of these flipper probes are compatible with tension imaging in live cells by fluorescence lifetime imaging microscopy.[2b]



General structure of papillon probes. From left to right: micelles in water, monomers in S_o and L_d states, and bulk membranes.

Deplanarized polycyclic aromatic compounds have attracted much attention since decades due to their unique spectroscopic, electrochemical and functional supramolecular properties.[3] The recently introduced *N,N'*-diphenyl-dihydrodibenzo[a,c]-phenazines[4] showed viscosity-sensitive and polarity-insensitive dual emission which originates from bent and planar excited states, preserving the same bent ground state. Inspired by this finding, we designed mechanophores[5] which undergo different excited-state conformational changes depending on environment, like butterflies or 'papillons' opening wings to fly (Figure). Upon comparing the emission ratio between planar and bent forms, clear differences were shown between micelles in water and monomers in solid-ordered (S_o) and liquid-disordered (L_d) lipid bilayer and in bulk membranes. Since their absorption maxima are located on relatively low wavelength for confocal laser scanning microscopy, TPEFM was chosen as an alternative technique for bioimaging.

Ongoing research is focused on the improving spectra-optical properties of the papillon probes to make them compatible with different imaging techniques and reveal forces at work in living systems.

[1] A. S. Klymchenko, *Acc. Chem. Res.* **2017**, *50*, 366.
[2] a) M. Dal Molin, Q. Verolet, A. Colom, R. Letrun, E. Derivery, M. Gonzalez-Gaitan, E. Vauthey, A. Roux, N. Sakai, S. Matile, *J. Am. Chem. Soc.* **2015**, *137*, 568; b) S. Soleimanpour, A. Colom, E. Derivery, M. Gonzalez-Gaitan, A. Roux, N. Sakai, S. Matile, *Chem. Commun.* **2016**, *52*, 14450.
[3] M. Rickhaus, M. Mayor, M. Juricek, *Chem. Soc. Rev.* **2017**, *46*, 1643.
[4] a) Z. Zhang, Y. Wu, K.-C. Tang, C.-L. Chen, J.-W. Ho, J. Su, H. Tian, P.-T. Chou, *J. Am. Chem. Soc.* **2015**, *137*, 8509; b) W. Chen, C.-L. Chen, Z. Zhang, Y.-A. Chen, W.-C. Chao, J. Su, H. Tian, P.-T. Chou, *J. Am. Chem. Soc.* **2017**, *139*, 1636.
[5] H. V. Humeniuk, A. Rosspeintner, G. Licari, V. Kilin, L. Bonacina, E. Vauthey, N. Sakai, S. Matile, *Angew. Chem. Int. Ed.* **2018**, 10.1002/anie.201804662; *Angew. Chem.* **2018**, 10.1002/ange.201804662.

Future plans

After my graduation, I started to pursue a PhD degree at the University of Geneva under the supervision of Professor Stefan Matile. I will continue my research on mechanosensitive membrane probes.

Zlatko Jončev
Nationality: *Serbian*
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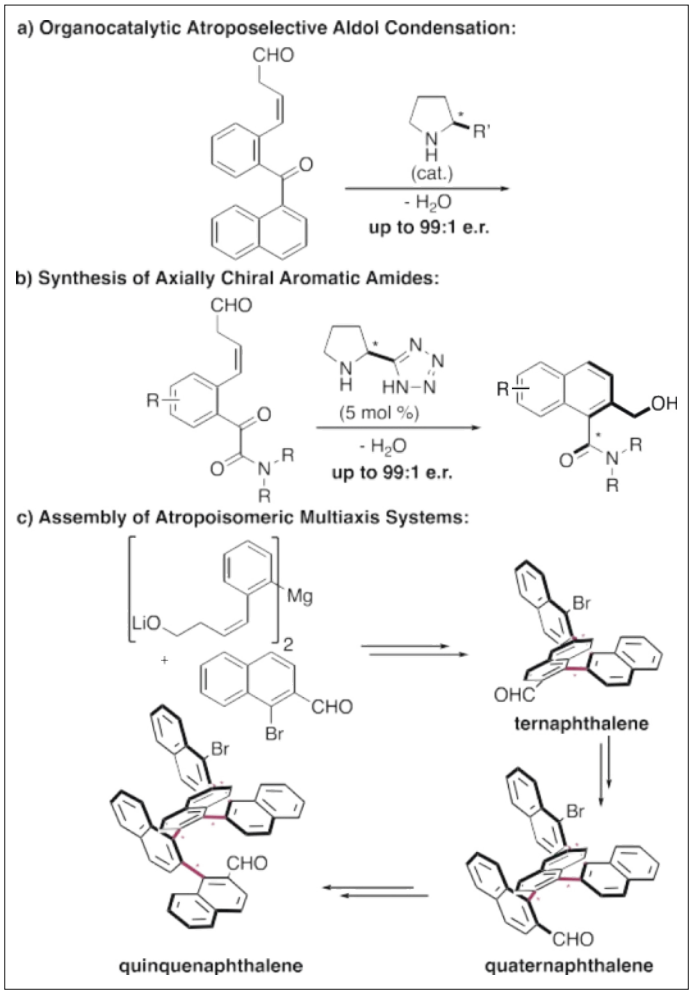
Master at: *University of Basel*
Master thesis supervisor:
Prof. Christof Sparr

Catalyst-controlled Atroposelective Arene Formation

Axially chiral biaryls are widespread among natural products, biologically active compounds and ligands for transition-metal catalysis. Despite their presence in various fields of chemistry, the selective formation of atropisomers remains challenging and the development of new stereoselective methods is highly desirable. Therefore, the focus of my Master thesis was on the discovery of new catalyst-controlled methods for the preparation of axially chiral biaryls.

Atropisomers are stereoisomers that arise from the restricted rotation about a single bond. Applications across different disciplines such as stereoselective catalysis, natural product research and medicine have made these compounds indispensable. Progress in different stereoselective methods to construct axially chiral biaryls, amongst others, includes cross-couplings of aryls, kinetic resolution, desymmetrization and [2+2+2]-cyclotrimerization. Often, catalyst-controlled atroposelective arene-construction requires transition metals that can be toxic in pharmaceuticals, even in trace amounts. Inspired by the biosynthesis of aromatic polyketides, our group is focusing on the organocatalytic atroposelective arene-forming aldol condensation. A *de novo* construction of arenes that simultaneously leads to a rotationally restricted aryl-aryl bond in a stereocontrolled fashion, has been developed to transform unsaturated ξ-ketoaldehydes to tri-*ortho*-substituted binaphthalenes with high atroposelectivities and yields (Scheme 1a).[1a]

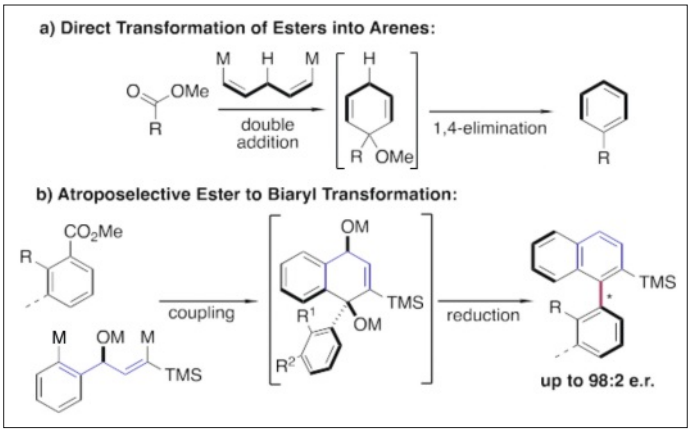


Scheme 1. Atroposelective Arene-Forming Aldol Condensation

The generality of this conceptually distinct methodology was confirmed in the study of axially chiral aromatic amides, a relevant class of molecules for medicinal chemistry.[1b] Arene formation from an α-ketoamide substrate was achieved by the addition of

a pyrrolidinyl tetrazole catalyst to form the desired rotationally restricted aromatic amide (Scheme 1b). With a subsequent *in situ* reduction, hydroxyl derivatives are obtained with high yields and exceptional atroposelectivities. Arene-forming aldol condensation enabled stereodivergent atropodiastereoselective synthesis, assembly of remarkable oligomeric compounds with more than one configurationally stable axis (Scheme 1c).[1c,d] Catalyst-controlled synthesis of ter-, quater- and quinquenaphtalenes allows a predictable spacial orientation of groups that is fundamental for the design of functional molecular systems.

A direct ester to arene transformation (dEAT) strategy was developed in our lab using 1,5-bifunctional organomagnesium reagent (Scheme 2a). Based on the dEAT, an atroposelective version was established that allowed for central-to-axial chirality conversion (Scheme 2b).[2] Initial coupling is followed by a



Scheme 2. Direct Ester to Arene Transformation (dEAT)

diastereoselective cyclization and an *in situ* reduction to directly obtain axially chiral biaryls with high selectivities.

These versatile synthetic concepts provide new reliable methods for the synthesis of various rotationally restricted compounds. The aldol condensation as well as the ester to arene transformation are conducted under mild conditions that allow for the preparation of a broad range of functionally distinct atropisomers. Ongoing studies focus on the application of defined molecular frameworks and advancement of new catalytic methods.

[1] a) A. Link, C. Sparr, *Angew. Chem. Int. Ed.* **2014**, *53*, 5458; *Angew. Chem.* **2014**, *126*, 5562; b) V. C. Fäseke, C. Sparr, *Angew. Chem. Int. Ed.* **2016**, *55*, 7261; *Angew. Chem.* **2016**, *128*, 7378; c) D. Lotter, M. Neuburger, M. Rickhaus, D. Häusinger, C. Sparr, *Angew. Chem. Int. Ed.* **2016**, *55*, 2920; *Angew. Chem.* **2016**, *128*, 2973; d) D. Lotter, A. Castrogiovanni, M. Neuburger, C. Sparr, *ACS Cent. Sci.* **2018**, *4*, 656.

[2] a) A. Link, C. Fischer, C. Sparr, *Angew. Chem. Int. Ed.* **2015**, *54*, 12163; *Angew. Chem.* **2015**, *127*, 12331; b) A. Link, C. Sparr, *Angew. Chem. Int. Ed.* **2018**, *57*, 7136; *Angew. Chem.* **2018**, *130*, 7254.

Future plans

I had the opportunity to continue my research in the group of Prof. Christof Sparr as a PhD student. My focus is on the development of new catalytic methods for the synthesis of rotationally restricted compounds.

Viktoriia Morad
Nationality: *Ukraine*
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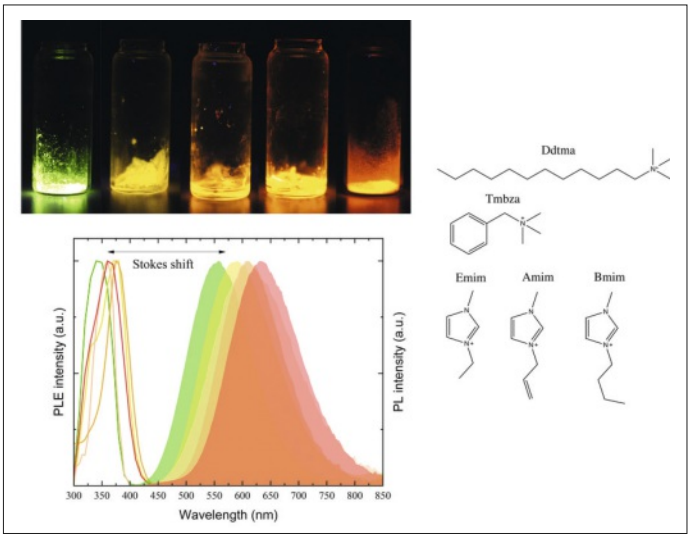
Master at: *ETH Zurich*
Master thesis supervisor:
Prof. Maksym V. Kovalenko

Exploration of Luminescent Hybrid Metal Halides

It is known for simple 3D connected metal halides that local lattice distortions lead to self-trapping of the free excitons. In general, lowering the dimensionality of the system and confining excitons to a single metal-halide center enhances self-trapping. The self-trapping leads to long lifetime of the luminescence and quantum yields close to 100%. The low-dimensional metal halide complexes were realized by using bulky organic cations for s^2-elements, e.g. Sb(III), Sn(II), Sb(II). Moreover, the tuning of optical properties can be achieved by the choice of the size and symmetry of the cation. The investigated complexes are of interest for applications in white-light emitting diodes, display technology, in energy technology as a down-conversion solar concentrators, as well as in biomedical research as fluorescent markers and thermography agents.

Optical properties, of low-dimensional metal halide complexes are strongly dependent on the local environment of the metal ion as the excitons are highly localized to the metal-halide center.[1,2] For this study, s^2-metal complexes were of interest. For example, broadband red emitters with extremely large Stokes shifts (1.6–1.7 eV) were synthesized based on Sn(II) and Sb(III) halides. It was discovered that the length and symmetry of the organic cation influences the optical properties. Crystal structure analysis revealed that organic cation influences overall symmetry of the metal-halide center. Since the Stokes shift is determined by the structural change of the metal-halide center upon excitation with ultra-violet light, more symmetric metal-halide centers demonstrate smaller Stokes shifts. Using this knowledge, the crystal structure of the metal-halide complexes can be designed in accordance with the desired emission color and Stokes shift.



Due to the low-dimensional nature of metal-halide complexes, they display strong temperature dependency of the emission lifetime. Typical emission lifetimes for the investigated Sb(III) complexes vary from thousands to tenths of nanoseconds within

the range from liquid N_2 temperature to 300 ºC. This enables the use of such complexes in thermometry.

In attempt to acquire the tool for facile structure tuning, the boundaries of the organic cations were explored, demonstrating that if very big and asymmetric organic cations with substituent chains longer than eight carbon atoms are used, crystallization does not occur and compounds stay in the state of frustrated ionic liquid at room temperature. In addition, such ionic liquid metal complexes containing Pb(II) display efficient luminescence, which has never been shown before for s^2-metal ionic liquids.

One of the other advantages of the discovered complexes is their easy processability due to low melting temperatures and facile synthesis from organic solvents.

[1] C. Zhou, Y. Tian, M. Wang, A. Rose, T. Besara, N. K. Doyle, Z. Yuan, J. C. Wang, R. Clark, Y. Hu, T. Siegrist, S. Lin, B. Ma, *Angew. Chem. Int. Ed.* **2017**, *56*, 9018.
[2] C. Zhou, Y. Tian, O. Khabou, M. Worku, Y. Zhou, J. Hurley, H. Lin, B. Ma, *ACS Appl. Mater. Interf.* **2017**, *9*, 40446.

Future plans

I have started a PhD in Prof. Kovalenko's Functional Inorganic Laboratory at ETH Zurich. My interest lies in the field of functional inorganic materials for energy applications. One of the goals of the field is to research sustainable and green technologies. On the other hand, I am fascinated by the fundamental research in the field of optical properties of inorganic materials.

Benjamin Planterose Jiménez
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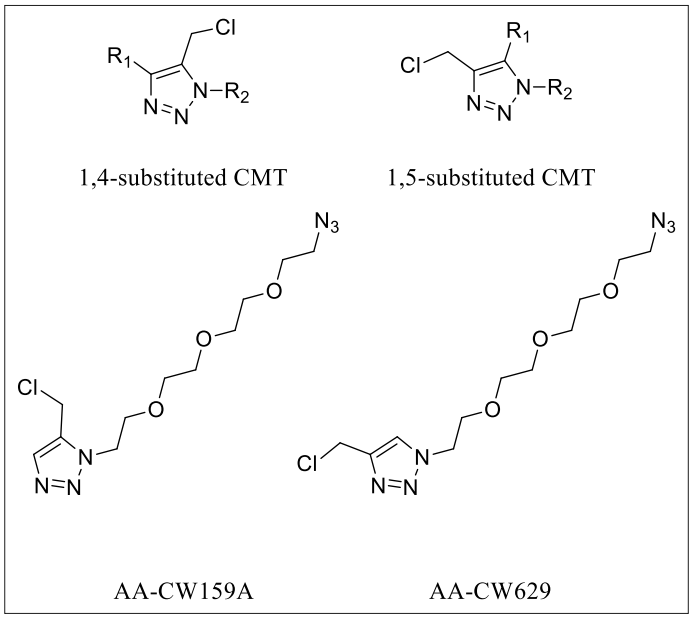
Bachelor at: *University of Sevilla*
Master at: *Université de Genève*
Master thesis supervisor:
Prof. Alexander Adibekian

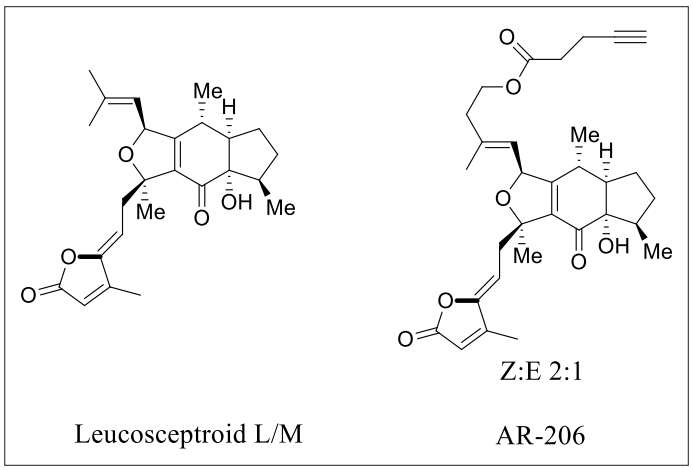
Synthetic and Natural Product-Based Covalent Drug Discovery: Chloromethyl Triazole Screening and the Bioprospecting of Leucosceptroid L/M

Synthetic and natural products have had profound implications for covalent drug discovery. In this project, both types of leads were explored. Chloromethyl triazoles are a class of highly-tunable easily-accessible cysteine-reactive compounds.[1] Additionally, leucosceptroids are a collection of antifeedant sesterterpenoid natural products derived from the Indochinese plant Leucosceptrum canum, some of which contain 1,4- and 1,6-conjugated systems that are also expected to form covalent adducts with protein cysteine residues.[2]

An already available probe of AA-CW159A mimicking the chloromethyl triazole (CMT) backbone was employed for chemoproteomics experiments. A broad range of human proteome targets with clinical interest reacted with the probe. Carrying out *in-gel* labelling experiments, binding to both probes AA-CW159A and AA-CW629 was confirmed for two key cell cycle regulators, CDK2 and CDK4 and for the downstream mTOR pathway protein, S6K1. All three targets were screened against 1,4- and 1,5-substituted CMT libraries employing a probe competition-based approach for which no hits were attained. CMT-based covalent leads are still an open endeavor. Future plans might focus on carrying out enrichment proteomics employing the probe AA-CW629 as well as expanding the size of the 1,4- and 1,5-substituted CMT libraries.



As part of a bioprospecting collaboration with Prof. Thomas Magauer's group (LMU Munich), we were supplied with total synthesis-derived leucosceptroid natural products and the corresponding functionalized probes. We performed leucosceptroid human protein target deconvolution by carrying out competitive proteomics profiling experiments with human lysates. Among leucosceptroid L/M targets we encountered BRAT1, a BRCA1 and mTOR interaction partner, involved in DNA damage response and cell proliferation. Increasing leucosceptroid L/M concentrations outcompeted BRAT1 probe labelling with an *in situ* IC_{50} of 97.5 μM. Motivated by this result, we devised functional assays to check whether protein function was impaired upon compound binding. Phosphorylated H2AX levels were not significantly changed upon treatment with leucosceptroid L/M suggesting that BRAT1 DNA-damage signaling functions remained invariant. Assessed by a plate scratching assay, we showed that cellular migration speed was not significantly altered when treated with the compound. Finally, immunocytofluorescence-based confocal microscopy revealed that BRAT1 subcellular localization was not jeopardized upon treatment with leucosceptroid L/M. Overall, we were unable to show function impairment upon binding. Further scaffold optimization is of deterrent difficulty due to the challenging synthesis required to access the product architecture. Instead, we would rather recommend the use of other more convenient previously reported BRAT1-binding structures such as curcumin, sulforaphane or zerumbone.



[1] C. Wang, D. Abegg, D. G. Hoch, A. Adibekian, *Angew. Chem. Int. Ed.* **2016**, *55*, 2911.
[2] C. L. Hugelshofer, T. Magauer, *Angew. Chem. Int. Ed.* **2014**, 53, 11351.

Future plans

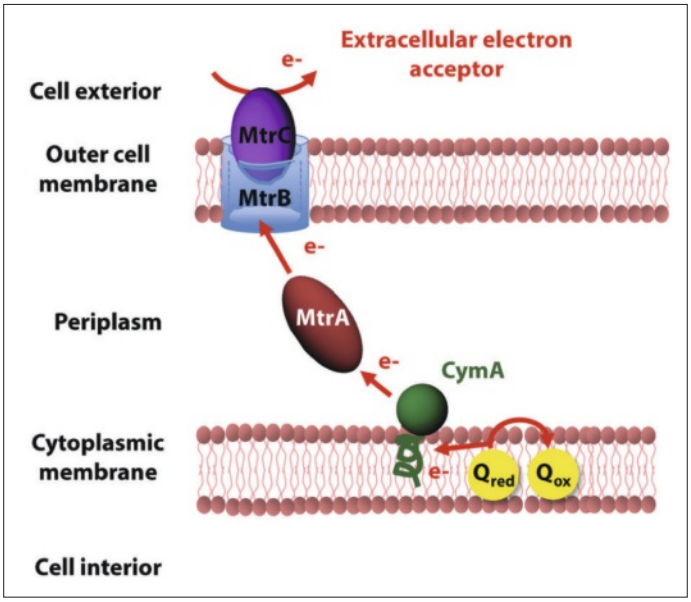
Prof. A. Adibekian, director of my Master Thesis, moved to the Florida campus of Scripps Research last October. I am currently performing an internship in the laboratory of Prof. Manfred Kayser's at the Department of Genetic Identification of the Erasmus MC in Rotterdam. I plan on starting my PhD studies at the beginning of next year.

Zahra Pourmand Tehrani
Nationality: *Iran*
Bachelor at: *Tehran Polytechnic*
Master at: *EPFL*
Master thesis supervisor:
Prof. Ardemis Boghossian

A Synthetic Biology Approach for Tuning an Extracellular Electron Transfer in *Escherichia coli*

Extracellular electron transfer is a specialized pathway in dissimilatory metal-reducing bacteria enabling efficient electron transfer between living microorganisms and external electron acceptors. Engineering bacteria with extracellular electron transfer capability overcoming the electrical insulation of the inner membrane can create a platform with the potential to be used in different technologies, including microbial fuel cells. Through introducing a mutation in one of the proteins in the electron conduit pathway, electron transport in Escherichia coli (E. coli) can be enhanced.

The focus of my thesis was on characterizing the heterologous expression of mutant variants of the MtrCAB pathway engineered in *E. coli*. This pathway is based on a network of multiheme c-type cytochromes of the dissimilatory metal-reducing bacterium *Shewanella oneidensis* MR-1, as shown in the Figure. The electron transfer between membrane-bound proteins in *E. coli* and the c-type cytochrome MtrA expressed in the periplasm of the bacteria is one of the main rate-limiting steps of this pathway.



Extracellular electron transfer pathway in Shewanella oneidensis MR-1[1]

In this work, the expression of *mtrA* variants generated by error-prone PCR was compared to the expression of wild type *mtrA* to ensure the selected mutations did not influence protein expression. Different factors were screened to optimize MtrC and MtrA expression in *E. coli*. The electron transfer capacity of the *E. coli* C43(DE3) *mtrCAB* strains was investigated through a medium-throughout colorimetric assay, which showed a higher extracellular electron transfer capacity in *mtrCAB* strains compared to strains without the MtrCAB pathway. However, observed differences in protein expression and extracellular electron transfer between the wild type and mutated versions of the *mtrCAB* strains must be further investigated to determine the role of *mtrA* mutation on functionality of the pathway.

[1] N. Schuergers, C. Werlang, C. M. Ajo-Franklin, A. A. Boghossian, *Ener. Environ. Sci.* **2017**, *10*, 1102.

Future plans

After graduation, I will pursue a PhD in the group of Prof. Thomas Lippert at the Paul Scherrer Institute and ETH Zurich. I am looking forward to further develop my skills for a successful career in academic or industrial research.