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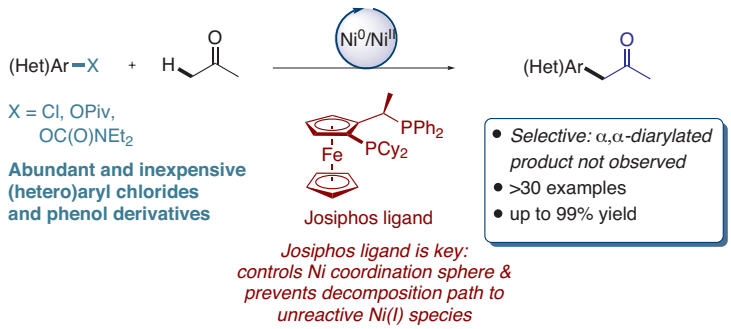
2021/03

Nickel-Catalyzed Mono-Selective α-Arylation of Acetone with Aryl Chlorides and Phenol Derivatives

Highlighted article by S. Abou Derhamine, T. Krachko, N. Monteiro, G. Pilet, J. Schranck, A. Tlili, A. Amgoune



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Dear Readers,

The pandemic has dramatically slowed down the research activity of most chemistry labs worldwide (including my own), and the effects are continuing. I am particularly worried for postgraduate research students and postdocs, who are being disproportionately hit and penalised by this situation. If I had to go through an entire year – or longer – of much-reduced research activity at that critically important time, my entire career would have been affected, delayed, and perhaps I would even have missed key milestones, such as securing my first independent position. I hope governments and research agencies will step in, putting in place effective ways to support the careers of early-stage researchers (ESRs), but we all need to prioritise their professional needs over everything else and be acutely aware of the consequences – even in terms of mental health – that this emergency will have on the future professional development and job opportunities of the younger generation, especially for those ESRs aiming to pursue a career in research.

The first article of this March 2021 issue of SYNFORM covers a very important aspect in drug discovery and development, namely the synthesis of deuterated *N*-alkyl pharmaceuticals enabled by a novel photocatalytic activation of water and alkanols discovered by Chenliang Su (P. R. of China). The second article takes us to the lab of A. Tlili and A. Amgoune (France) who discovered that aryl chlorides and phenol derivatives can be used to achieve the challenging mono-arylation of acetone via nickel catalysis. The third Literature Coverage article welcomes back the art of total synthesis, and specifically that of the cyclodepsipeptide alveolaride C, whose structural assignment was also investigated by R. K. Goswami (India). The final article is an interview with a new member of the Editorial Board of the Thieme-Chemistry Journal ORGANIC MATERIALS, namely Professor Michael Mastalerz (Germany).

Enjoy your reading!!

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Semiconductor Photoredox Catalysis to Engineering Deuterated *N*-Alkyl Pharmaceuticals

Nat. Commun. **2020**, *11*, 4722

'Deuterium switch' in therapeutic molecules is widely used to study and alter the absorption, distribution, metabolism and excretion of pharmaceuticals. In 2017, the first deuterium-labeled drug, deutetrabenazine, was approved by the FDA, ushering a new era of deuterated clinical drug development. "Among the myriad of commercial drugs, over 50% of the top-selling drugs contain *N*-alkyl amine units and *N*-dealkylation by cytochrome P450 (CYP450) is a very common metabolic pathway in such *N*-alkyl drugs," said Professor Chenliang Su (Shenzhen University, P. R. of China). "Thus, deuterium substitution of *N*-alkyl groups in *N*-alkyl drug molecules could impact their pharmacodynamic properties by slowing down the N–C bond cleavage. In this regard, the precision synthesis of drug analogues with deuterated *N*-alkyl amine units holds great promise but is still a challenging endeavor from the chemistry viewpoint." Traditional approaches to these *N*-alkyl drugs generally rely on N-alkylation with deuterated alkyl halides such as CD_3I or reduction of N–CO_2R moieties with $LiAlD_4$. "The substitution is of interest as these deuterated reagents are often highly toxic, carcinogenic and/or volatile, the latter generally causing high costs and waste production," added Professor Su.

Professor Su's group had previously developed a controllable deuteration of halides and alkenes via semiconductor-promoted photocatalytic D_2O splitting (*Nat. Commun.* **2018**, *9*, 80; *Adv. Sci.* **2019**, *6*, 1801403). In this new article, they developed a groundbreaking semiconductor photoredox catalysis to achieve mild and controllable deuterium-labeling in N-alkylated pharmaceuticals via D_2O splitting coupled with isotopic alkanol oxidation. "With the goal of achieving controllable isotope-labeling in N-alkylated amines, in this work we rationally designed photocatalytic water-splitting to furnish [H] or [D] by photogenerated electron-induced reduction; meanwhile, photogenerated electron-holes with appropriate oxidative ability are utilized to selectively oxidize isotopically labelled alkanols, furnishing the corresponding aldehydes for aldehyde-amine condensation to afford isotopically labelled imine intermediates. These imines could be subsequently reduced by [H]/[D] from water splitting, producing the corresponding *N*-alkyl chemicals and drugs," said Professor Su, who continued: "Benefitting from this unique design, low-cost and sustainable isotopic water and alkanols are proposed as a combined deuterated alkylation reagent for the first time. More importantly, precise control of the number of deuterium atoms (i.e., *N*-CD_3, *N*-CD_2H and *N*-CDH_2) at the potential metabolic position of *N*-methyl drugs is enabled by simply tuning the deuteration of isotopic water and methanol." Gratifyingly, the group found that this photocatalytic strategy exhibited a broad reaction scope, good functional group tolerance, high selectivity and excellent deuterium incorporation. "Substrates including primary amines, secondary amines, amino acid derivatives and heterocyclic amines readily underwent N-trideuteromethylation reactions, furnishing the corresponding products with high deuterium incorporation (up to 98%) and excellent yields (up to 94%). Sensitive substrates with alkyl chiral centers were compatible and unperturbed," explained Professor Su. "Late-stage functionalization of various commercial pharmaceuticals such as flutamide, nimesulide, fluoxetine, tetracaine, atomoxetine, sertraline, paroxetine and vortioxetine was successfully demonstrated. Impressively, this mild and general process enables access to site-specifically labeled drugs in a single step," he added. Professor Su continued by explaining that deuterated *N*-alkyl pharmaceuticals, including imipramine-d_3, loxapine-d_3, alverine-d_5 and dofetilide-d_3, were successfully obtained and gram-scale synthesis could be easily achieved. "Last but not least, this protocol has been nicely applied for the facile synthesis of *N*-CD_3, *N*-CD_2H and *N*-CDH_2 nimesulide derivatives, butenafines-d_3, d_2 and d_1, loxapines-d_3, d_2 and d_1 and imipramines-d_3, d_2 and d_1, with high yields and uniformly high D-incorporation (> 95%)," he said.

Professor Su concluded: "This study not only paves the way to the precision deuterium-labeling at potential metabolic sites of *N*-alkyl pharmaceuticals, which may provide a reliable shortcut for the discovery of new deuterated drugs, but also reveals the potential of semiconductor photocatalysts in artificial photosynthesis of pharmaceuticals with water and organics."

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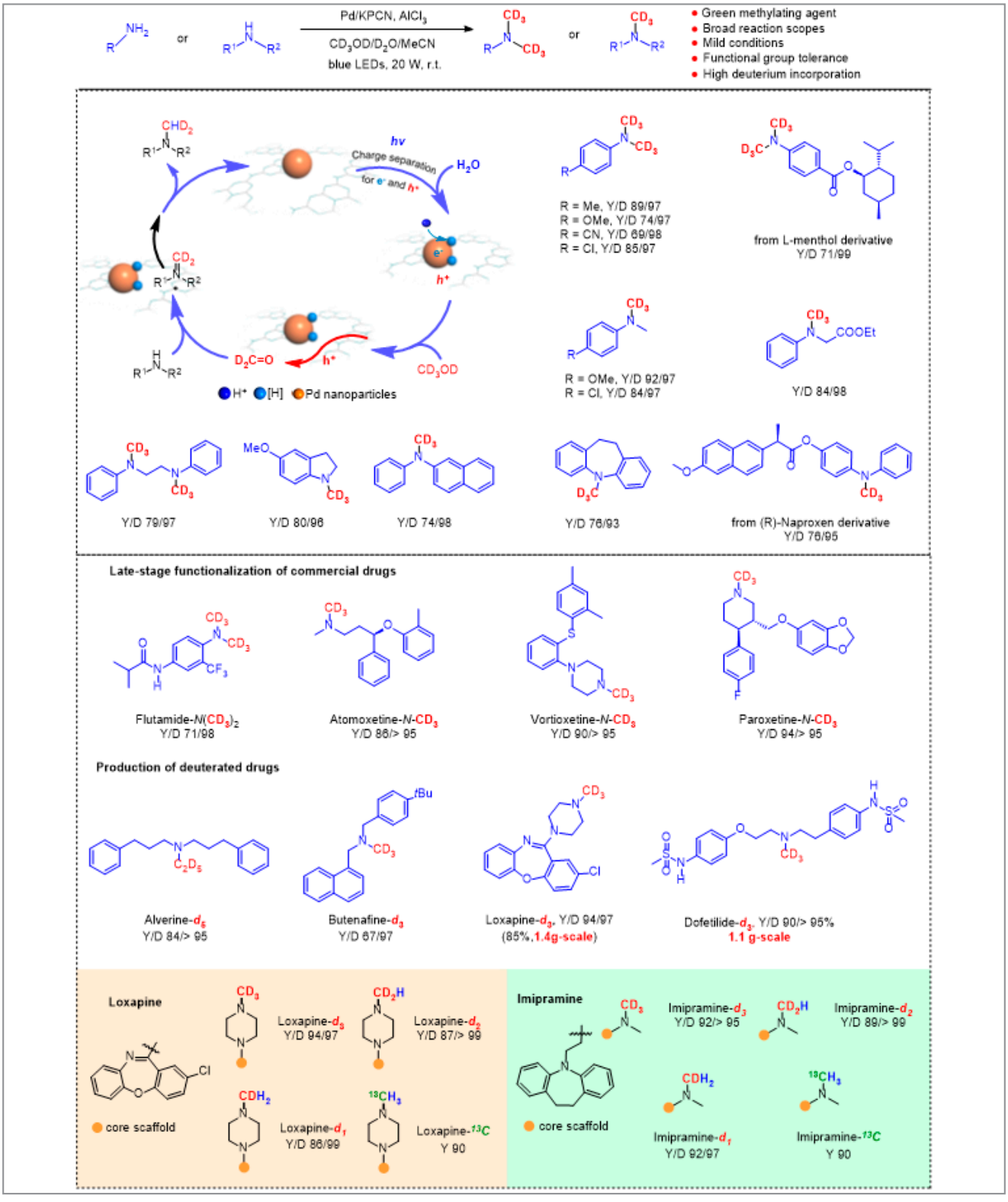

Scheme 1 Selected examples of controllable D-labeled N-alkylation of pharmaceutical-related amines by semiconductor photoredox catalysis

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About the authors

Dr. Z. Zhang

Zhaofei Zhang received his M.S. degree from the Department of Chemistry, Zhengzhou University (P. R. of China) in 2015, and obtained his Ph.D. from the Institute of Chemistry, Chinese Academy of Sciences (P. R. of China) in 2018. He is a postdoctoral researcher at the International Collaborative Laboratory of 2D Materials for Optoelectronics Science and Technology (ICL-2D MOST), Shenzhen University (P. R. of China). His current research interest includes developing synthetic methodologies of semiconductor photocatalysis.

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Prof. C. Su

Chenliang Su received his BS degree (2005) and Ph.D. (2010) from the Department of Chemistry at the Zhejiang University of China (P. R. of China). After that he worked as a research fellow at the Advanced 2D Materials and Graphene Research Centre at the National University of Singapore (Singapore, 2010–2015). He is now a full professor at the International Collaborative Laboratory of 2D Materials for Optoelectronics Science and Technology (ICL-2D MOST), Shenzhen University (P. R. of China) and a Principal Investigator at the ICL-2D MOST in materials science. His current interests include the study of nanostructured materials for heterogeneous catalysis and energy applications.

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Nickel-Catalyzed Mono-Selective α-Arylation of Acetone with Aryl Chlorides and Phenol Derivatives

Angew. Chem. Int. Ed. **2020**, *59*, 18948–18953

The development of straightforward methods for preparing α-arylated ketones directly from acetone is of great interest from both economic and synthetic points of view. While the arylation of carbonyl compounds with transition metal catalysts, via the formation of enolate intermediates, represents a powerful and relatively well-established class of carbon–carbon bond-forming reactions,[1] the transposition of this technology to simple acetone has remained – in comparison – rather elusive.[2] For example, this challenging transformation has rarely been achieved using palladium catalysis with aryl halide electrophiles. "From an economic viewpoint, the development of this methodology with more sustainable and cost-effective nickel catalysts appeared as a very valuable alternative, but the transposition to nickel has proven highly challenging," remarked Professor Abderrahmane Amgoune, from CNRS & Université Lyon 1 (Villeurbanne, France). He continued: "Only a few examples of nickel-catalyzed arylations of substituted ketones were reported and the arylation of acetone had not been documented with nickel catalysts thus far."

This project emerged from collaborative work between the group of Professor Amgoune, colleagues Dr. Nuno Monteiro and Dr. Anis Tlili (all from Université de Lyon), and Dr. Johannes Schranck from Solvias AG (Switzerland). The story started with a discussion between Professor Amgoune, Dr. Tlili and Dr. Schranck on the potential of Josiphos-type ligands in nickel catalysis, especially for the functionalization of small molecules.[3] "We decided to combine my expertise in synthetic organometallic chemistry and mechanistic studies,[4] with the experience of Dr. Tlili and Dr. Schranck in transition-metal-catalyzed functionalization of small building-block molecules[5] to develop and understand the nickel-catalyzed monoarylation of acetone," said Professor Amgoune.

Dr. Tlili approached Professor Amgoune and Dr. Monteiro with a preliminary result showing that the combination of Josiphos ligand (see scheme for structure) with $Ni(COD)_2$ could promote the monoarylation of acetone, albeit in low yield. Professor Amgoune explained: "The collaboration started at that point, and beyond the synthetic interest of this transformation we were also eager to carry out detailed mechanistic studies to unravel the key role of the ligand."

"We started our investigations with intensive optimization studies that were carried out by graduate student Sary Abou Derhamine," explained Professor Amgoune, who continued: "He investigated a large set of reaction conditions and ligands and identified a very practical catalytic system that demonstrated a high compatibility with several functional groups and enabled the formation of complex substrate structures with good to excellent yields (Scheme 1)."

Even more important than its economic interest, nickel catalysts display reactivities that are complementary to those of palladium. In particular, the group took advantage of the ability of nickel to activate C–O bonds to extend the methodology to the unprecedented coupling of acetone with phenol derivatives.

While these optimization studies revealed that the association of Josiphos ligand (SL-J004) and $Ni(COD)_2$ was a very efficient catalytic system, they also raised several mechanistic questions about the key influence of the ancillary ligand. Strikingly, the researchers discovered that only Josiphos-type ligands were effective under these reaction conditions, moreover they also observed that a second equivalent of ligand was beneficial to the catalytic performance.

"We decided to carry out detailed mechanistic investigations to address these questions," said Professor Amgoune. He continued: "The mechanistic work – which combined NMR monitoring, isolation and characterization of key catalytic intermediates – was carried out principally by postdoctoral associate Dr. Tetiana Krachko, who succeeded in showing the feasibility of all mechanistic steps of the envisioned catalytic cycle." Professor Amgoune then went on to credit Dr. Guillaume Pilet with carrying out the X-ray diffraction analysis of isolated Ni(0) and Ni(II) intermediates. He remarked: "These important mechanistic studies revealed that the specific reactivity of (Josiphos)Ni catalyst likely results from an enhanced stabilization of the catalyst resting state Ni(II)-aryl species towards decomposition to unreactive Ni(I) species. We were also surprised to find that the use of an excess of Josiphos ligand was not deleterious for the catalytic reactivity. Indeed, under these conditions we could form a saturated 4-coordinate $Ni(bis\text{-}phosphine)_2$ species that was anticipated to be unreactive, based on previous reports. However, we have shown that the extra ligand may indeed coordinate to Ni(0) at high temperature to give a $Ni(Josiphos)_2$ species, which was characterized. We think that the steric

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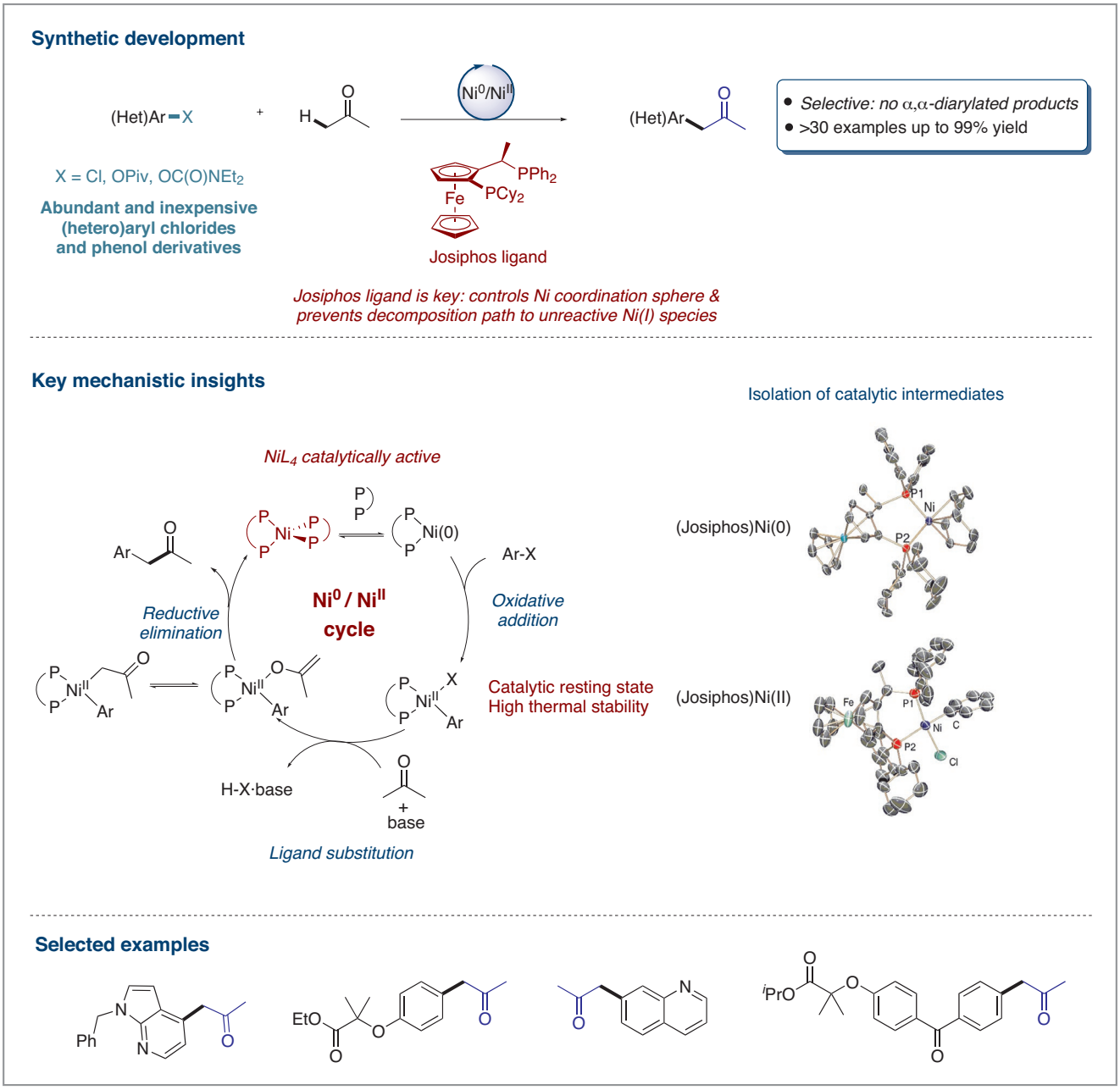
bulk of the Josiphos-type ligand, combined with the specific stereoelectronic properties imparted by the ferrocene backbone, renders the Ni(Josiphos)$_2$ very susceptible to ligand exchange with the aryl halide substrate."

Overall, important fundamental information on the unique behavior of Josiphos-type ligands with nickel emerged from this study. Professor Amgoune concluded: "We are currently seeking to further understand and harness the specific coordination properties of Josiphos-type ligands for the development of new nickel-catalyzed transformations."



Scheme 1 Nickel-catalyzed mono-arylation of acetone: synthetic development and mechanistic studies

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About the authors

Dr. S. Abou Derhamine

Sary Abou Derhamine was born in Kfarkouk (Lebanon) and graduated with distinction from the Lebanese University of Beirut (Hadath) with an M.Sc. degree in physical chemistry. Then he was awarded a scholarship for excellence in science from the French government to pursue a Master's degree in molecular chemistry and catalysis at the University of Lyon (France). After that, he carried out a multidisciplinary internship where he worked with physicists, chemists and engineers under the supervision of Prof. B. Masenelli, Dr. A. Quadrelli and Dr. J. Canivet. Subsequently, he completed his Ph.D. in 2020 working in the development of nickel-catalyzed cross-coupling reactions under the guidance of Prof. A. Amgouen and Dr. N. Monteiro at the University of Lyon. In November 2020, he started a postdoctoral position at Total Research & Technology Feluy (Belgium) in collaboration with Prof. Jean-Francois Carpentier (Rennes, France).

Dr. T. Krachko

Tetiana Krachko received her Ph.D. (2018) in organophosphorus chemistry from the University of Amsterdam (The Netherlands) under the supervision of Assoc. Prof. J. C. Slootweg. Subsequently, she conducted her postdoctoral research at the same university with Prof. J. N. H. Reek, working on the development of new self-assembled ligands for hydroformylation. Afterwards, she pursued her postdoctoral fellowship at the University of Lyon (France) with Prof. A. Amgoune, performing the synthesis of organometallic complexes and studying their reactivity for the activation and functionalization of inert bonds. She has been working as a research engineer at the chemical startup Fairbrics (France) since August 2020, focusing on the production of synthetic fibers from carbon dioxide.

Dr. N. Monteiro

Nuno Monteiro studied chemistry at the University of Lyon (France) and completed his Ph.D. there in 1992, working on the development of Pd-catalyzed Conia-ene-type cyclizations under the guidance of Jacques Goré and Geneviève Balme. Following a one-year period as a contractual lecturer, he joined the group of Varinder K. Aggarwal (University of Sheffield, UK) as a Marie Curie postdoctoral fellow to work on the synthesis of polyoxin analogues. In 1996 he returned to Lyon where he was appointed as a CNRS researcher. His main research interests have concerned the development of transition-metal-catalyzed methods toward the construction and functionalization of heterocycles, with a recent focus on organofluorine chemistry.

Dr. J. Schranck

Johannes Schranck obtained an M.Sc. in chemistry at the University of Rostock in northern Germany. He conducted his Ph.D. studies in the group of Prof. Matthias Beller at the Leibniz-Institute for Catalysis (Germany) as well as with Prof. Mark Stradiotto at Dalhousie University (Canada). In 2014, he joined the catalysis group of Solvias in Basel (Switzerland) to work on the development of industrial asymmetric hydrogenations and

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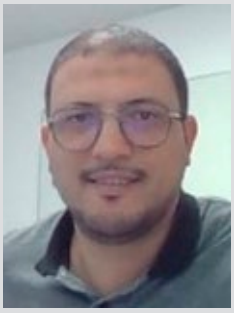
cross-coupling reactions. Since 2019, Johannes has been leading Johnson Matthey's homogeneous catalysis team as R&D Manager in West Deptford, NJ (USA). In this role he is focused on the development and commercialization of homogeneous catalysis solutions for the global pharmaceutical and fine chemical industry.



Dr. A. Tlili

Anis Tlili is CNRS Research Fellow. After gaining a Master's degree with Prof. S. Jugé (Dijon, France), he completed his Ph.D. in the group of Dr. M. Taillefer (Montpellier, France) in late 2011. Subsequently, he joined the group of Prof. M. Beller (LIKAT, Rostock Germany) for a postdoctoral stay before taking up a second collaborative postdoctoral position (CEA/ICSN, Paris). In 2014 he was appointed a CNRS research fellow in Lyon. His current research interests lie in the area of homogeneous catalysis/photoredox catalysis, with particular focus on fluorine chemistry. Anis was recently awarded a JSP Fellowship for the 53rd Bürgenstock Conference (2018) and was selected to participate to EuCheMS Young Investigators Workshop in Oxford in 2018.

Prof. A. Amgoune

Abderrahmane Amgoune received his PhD in 2006 at the University of Rennes (France) under the supervision of Profs. J.-F. Carpentier and C. Thomas. He then moved to the University of Konstanz (Germany) as an Alexander-von-Humboldt postdoctoral research associate with Prof. S. Mecking. In 2008, he was appointed a CNRS research position at the University of Toulouse (France). In 2017, he was promoted to full professor at the University of Lyon (France). His research interests range from fundamental organometallic chemistry to the development of dual catalytic strategies with transition metals for the functionalization of inert bonds. His work has received several awards, including most recently a nomination as junior member at the Institut Universitaire de France (2019), the Thieme Chemistry Journals Award (2020) and the Young Investigator Award from the French Chemical Society, Organic Chemistry Division (2016).

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Cyclodepsipeptide Alveolaride C: Total Synthesis and Structural Assignment

Chem. Sci. **2020**, *11*, 11259–11265

Alveolarides are a class of agriculturally important cyclodepsipeptides, which were isolated from cultures of the fungus *Microascus alveolaris* in 2018 by researchers at Dow AgroSciences (DAS) in the USA who partially elucidated the structures of alveolarides A–C (*J. Nat. Prod.* **2018**, *81*, 10–15). These are 17-membered macrocyclic compounds having the rarely found 2,3-dihydroxy-4-methyltetradecanoic acid (DHMTDA) as the common nonpeptide unit. The stereochemistry of asymmetric centers on the DHMTDA segment remained unassigned. The group of Professor Rajib Kumar Goswami at the Indian Association for the Cultivation of Science (Jadavpur, India) has targeted alveolaride C (Figure 1) with the intention of solving the mystery of its structural assignment using total synthesis as the key tool. Professor Goswami remarked: "There are three asymmetric centers in the DHMTDA fragment of alveolaride C, whose configuration is unknown, giving rise to the possibility of eight configurational isomers (Figure 2); finding the actual structure of alveolaride C was quite challenging."

A convergent synthetic route was adopted by Professor Goswami and his co-workers. To narrow down the search for structural possibilities, four isomers (**1a–d**) (Figure 2) of the DHMTDA fragment were synthesized initially and their data were compared with the reported data. "The data of isomer **1d** matched nicely with the reported one," explained Professor Goswami. He continued: "However, isomer *ent*-**1d** was also taken into account, as no specific rotational value of the DHMTDA fragment was reported by the DAS team." According to Professor Goswami, selective esterification of a β-hydroxyl group in the presence of a free α-hydroxyl on the key D-serine-DHMTDA synthetic intermediate (first step in Scheme 1) was one of the crucial steps, which was likely possible due to the presence of hydrogen bonding between the α-hydroxyl and the amide carbonyl. This possibility was conceived from the crystal structure of isomer *ent*-**1d**. "Two possible structures (**2a** and **2b**) of alveolaride C were synthesized from DHMTDA counterparts **1d** and *ent*-**1d** and their NMR spectroscopic data were compared with the reported data of the isolated alveolaride C," said Professor Goswami. He continued: "The isomer **2b**, having 29*S*,30*R*,31*S* configuration of the DHMTDA unit, was close to the reported NMR data of isolated alveolaride C. However, a few noticeable discrepancies in the ^{1}H NMR data were observed for the protons nearby the β-phenylalanine and the DHMTDA counterpart." This observation led the group to

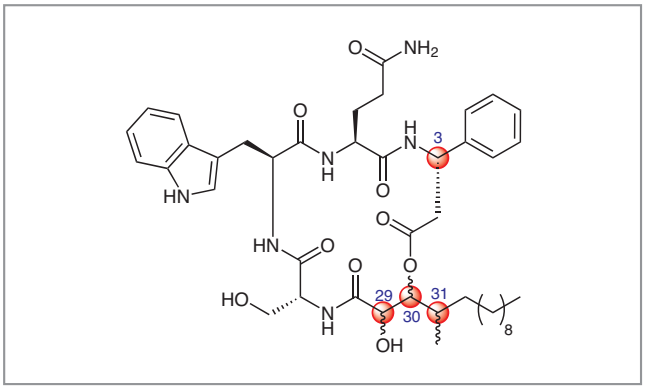

Figure 1 Putative structure of alveolaride C

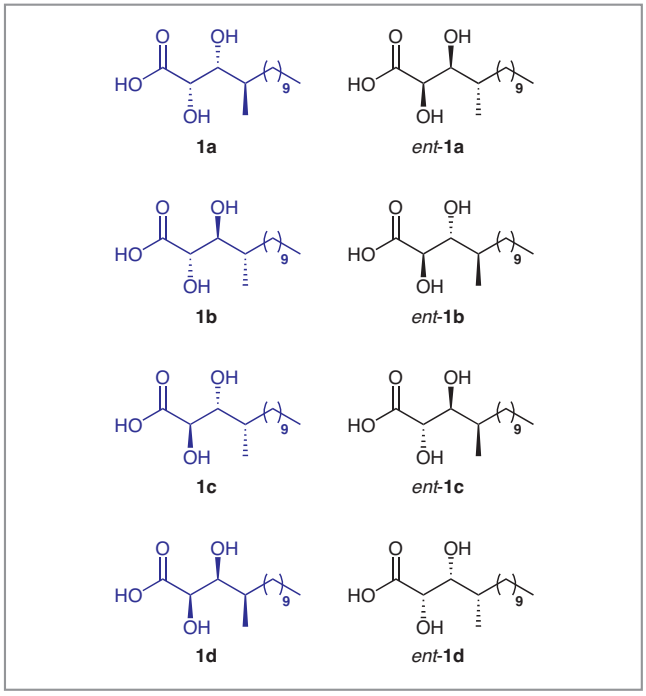

Figure 2 Possible stereoisomers of DHMTDA (**1a–d** and *ent*-**1a–d**)

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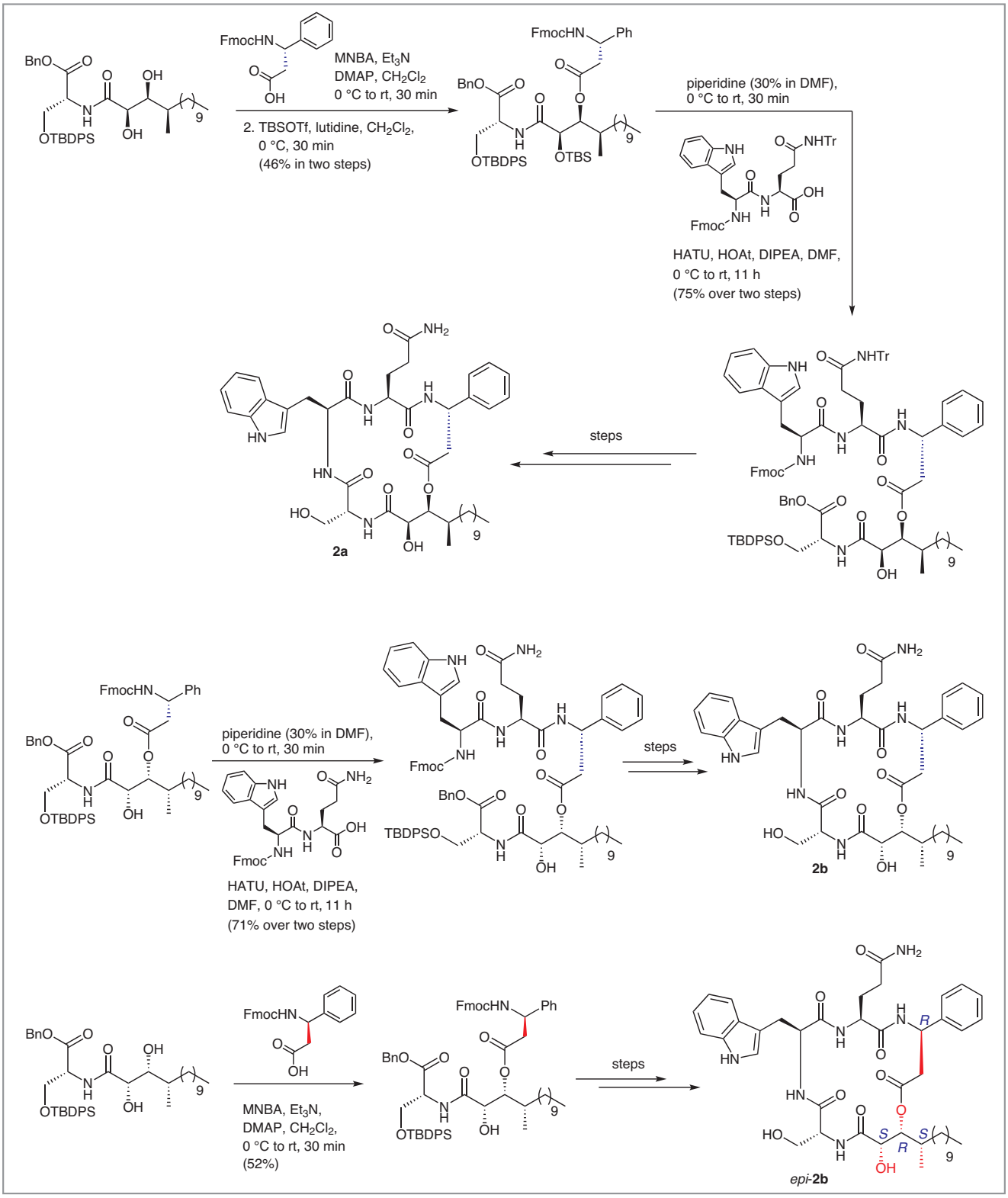

Scheme 1 Total synthesis of alveolaride C and two of its diastereoisomers

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reconsider the absolute configuration of the β-phenylalanine fragment. Thus, the group synthesized the C-3 epimer (*epi*-**2b**) of the compound **2b** which turned out to be in complete accordance with the data of the isolated natural product.

"The structural riddle of alveolaride C was solved successfully," said Prof Goswami. "The absolute stereochemistry of three undetermined centres on the DHMTDA segment was established unambiguously as 29*S*,30*R*,31*S*. The stereochemistry of the β-phenylalanine unit was revised from *S* to *R*." Professor Goswami concluded: "This synthetic study was crucial for determining the unassigned stereocenters common to all the members of this family and also a very important stepping-stone for the total synthesis of other members of this family."

About the authors

Prof. R. K. Goswami

Rajib Kumar Goswami obtained both his B.Sc. and M.Sc. degrees in chemistry from Calcutta University (India) in 1999 and 2001, respectively. In 2007, he completed his doctoral studies on chemical synthesis of natural products from Indian Institute of Chemical Technology, Hyderabad (India), under the supervision of Prof. Tushar K. Chakraborty. He was awarded his Ph.D. in chemistry by the University of Kalyani (India) in 2008. He moved to The Scripps Research Institute, San Diego (USA) in 2007 with a postdoctoral fellowship and worked with Prof. Subhash C. Sinha until 2011. He is currently a Professor in the School of Chemical Sciences, Indian Association for the Cultivation of Science, Kolkata, India. His research interests focus on asymmetric total synthesis of bioactive natural products and evaluation of their bio-medicinal applications.

S. Saha

Sanu Saha received his B.Sc. and M.Sc. degrees in chemistry from Calcutta and Jadavpur Universities (India) in 2015 and 2017, respectively. Currently he is working in the group of Prof. Goswami at the Indian Association for the Cultivation of Science in the School of Chemical Science. His research interest is the synthesis of natural products.

D. Paul

Debobrata Paul is pursuing his Ph.D. under the supervision of Prof. Goswami at the School of Chemical Sciences, Indian Association for the Cultivation of Science. He completed both his B.Sc. and M.Sc. degrees in chemistry from The University of Burdwan (India) in 2012 and 2014, respectively. He is currently working on the total synthesis of bioactive macrocyclic natural products.

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Editorial Board Focus: Professor Dr. Michael Mastalerz (Organisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, Germany)

Background and Purpose. From time to time, SYNFORM portraits Thieme Chemistry Editorial Board or Editorial Advisory Board members who answer several questions regarding their research interests and revealing their impressions and views on the developments in organic chemistry as a general research field. This Editorial Board Focus presents Professor Dr. Michael Mastalerz (Organisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, Germany) who is the Editor-in-Chief of ORGANIC MATERIALS, the new Thieme Open Access Journal launched in 2019.

Biographical Sketch

Prof. Dr. M. Mastalerz

Michael Mastalerz studied chemistry at the Gerhard-Mercator-Universität in Duisburg, Germany where he got his diploma in 2002. He then moved to the Ruhr-Universität Bochum (Germany) for his PhD studies, which he finished in 2005. After a short stay in industry, he carried out postdoctoral research in the group of Gregory C. Fu at MIT in Cambridge (USA) from 2006 to 2007. He returned to Germany for a second postdoctoral position at Ulm University to work in the group of Peter Bäuerle on dendritic oligothiophenes for organic electronic applications. At Ulm University he also started his independent career, working towards his habilitation which was successfully completed in February 2013. In April 2013 he became professor of organic chemistry at the Ruprecht-Karls-Universität Heidelberg (Germany). His main research interests focus on the synthesis of large shape-persistent organic cage compounds as soluble porous molecules, hydrogen-bonding based chiral capsules and large soluble fused contorted aromatic compounds. He is an ERC consolidators grant holder and Editor-in-Chief of the recently launched new Thieme journal ORGANIC MATERIALS.

INTERVIEW

SYNFORM *Please comment on your role as a member of the Editorial Board of ORGANIC MATERIALS.*

Prof. Dr. M. Mastalerz Although I am officially Editor-in-Chief, I do not see any hierarchical differentiation between Pol Besenius, Xiaozhang Zhu and me in their role as editors. I understand my role is to serve authors as scientists, in form of making the whole process from submission to publication as convenient and fair as possible, which starts with choosing the right reviewers, who are interested in the work but not biased or direct competitors. I believe that only reviewers who are interested in the content of a manuscript will deliver a strong and reliable review on time and do not see the whole process as a 'burden'. Currently, the reviewing process for ORGANIC MATERIALS has become very fast (the fastest peer-reviewing process I've seen, which included two comprehensive and useful reviews, took less than one day!) and we need to keep up this high standard.

Since we, the editors, are active researchers ourselves, we understand what authors expect from editors. Besides scientific fairness, it is the quality of the publications paired with short times from submission to publication.

In addition to the routine work as editors, we give authors the opportunity to contribute to virtual issues of special topics, which are currently 'hot topics' in the field. With this editorial tool, we create high visibility within specific communities.

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SYNFORM *How do you describe the value of a product such as ORGANIC MATERIALS to the chemistry community?*

Prof. Dr. M. Mastalerz The value of a journal such as ORGANIC MATERIALS is very high, because it brings a community closer together simply because the topics are narrower than in, for example, more general materials journals. On top of this, all content is open access, which allows everybody who is interested in the publications to download all for free, read and be stimulated for their own research – worldwide.

SYNFORM *What is the focus of your current research activities?*

Prof. Dr. M. Mastalerz Currently we are working on shape-persistent organic cage compounds of varying size, shape and function. The cage compounds are made mainly by imine condensation. This research topic was the basis of my independent career and indeed most of my co-workers still work on it. We are eager to understand formation mechanism in more detail (e.g. kinetic vs. thermodynamic control), as well as being able to structurally tune those cages for certain materials applications, such as selective gas sorption.

A few years ago, we also stepped into the field of fused polycyclic aromatic compounds (PACs) and hydrocarbons (PAHs). We initially found that triptycene end-capping can, for example, be used to control packing of larger π-systems or prevent aggregation if two or more of these units are present, making typically insoluble dyes and fused aromatics highly soluble. In this respect our triptycene end-capped hexabenzo-ovalene (HBO) has some very nice orange fluorescence and has been used in electrochemical light emitting cells, showing superior properties.

Furthermore, negatively curved PACs caught our attention. We synthesized the first conformational stable chiral monkey-saddle PAH or a bis-azulene based PAH building the basis for even more interesting systems. I promise there will be more 'fancy' molecules to come out of our lab!

SYNFORM *You are a leading researcher with regard to organic materials chemistry. Could you tell us more about how important you perceive this particular topic to be?*

Prof. Dr. M. Mastalerz I would not necessarily call myself a leading researcher, but yes, with our seminal papers on porous organic cages (POCs) we are among the pioneers who shaped the field and may still influence the community that is steadily growing. I am really happy to see that more and more, especially younger and talented groups from all over the world have very creative ideas to bring this topic forward. In the beginning, such porous organic cages could not compete with existing ones, such as metal-organic frameworks (MOFs) or covalent organic frameworks (COFs). Meanwhile, POCs are established materials and have found their place within the family of porous materials.

SYNFORM *What is your most important scientific achievement to date and why?*

Prof. Dr. M. Mastalerz In my opinion we have a few really important ones; probably the most important is our paper from 2011 (*Angew. Chem. Int. Ed.* **2011**, *50*, 1046–1051), demonstrating that porous materials with high specific surface areas can also rely on molecular crystals and not just networks. This paper is in the meantime highly cited. More importantly, based on these findings described therein I had a lot of invitations to speak about its content at conferences, symposia and institutional seminars, which is the greatest honor you can experience as a young researcher. This paper really accelerated my career.

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Coming soon

Literature Coverage

New Uses for Recycled Carbon: Converting Waste Polyethylene into Alkylaromatics

Literature Coverage

Mimicking Oxidative Radical Cyclizations of Lignan Biosynthesis Using Redox-Neutral Photocatalysis

Literature Coverage

Polyethylene Upcycling to Long-Chain Alkylaromatics by Tandem Hydrogenolysis/Aromatization

Further highlights

Synthesis **Review: Shining Light on the Light-Bearing Element: A Brief Review of Photomediated C–H Phosphorylation Reactions**
(by C.-J. Li and co-workers)

Synlett **Account: From Enantioselective to Regiodivergent Epoxide Opening and Radical Arylation – Useful or Just Interesting?**
(by A. Gansäuer)

Synfacts **Synfact of the Month in category "Polymer-Supported Synthesis": Photoredox C–O Cross Coupling of Aryl Halides and Alcohols on Nickel-Coordinated Carbon Nitride**

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