제 249 회 유기화학 세미나

Korean Chemical Society
Division of Organic Chemistry

PROGRAM BOOKLET

http://www.kcsorganic.org

일시: 2022 년 12 월 2 일
장소: 고려대학교 SK 미래관 최종현홀

공식후원사: TCI · SEJIN CI

프로그램 안내: 12/02 (Fri)

	Session I. 좌장: 이안나 (전북대)
13:30-13:40	**인사말 (김종승 대한화학회 유기화학분과회 회장, 고려대)**
13:40-14:00	**박정우 (목포대)** “Migratory Hydrofunctionalization of Alkynes Enabled by Cobalt Catalysis”
14:00-14:20	**장원준 (동아대)** “Oxidant-induced Direct α-Alkylation of Ketones Enabled by Ir-B Cooperative Catalysis”
14:20-14:40	**박윤수 (KAIST)** “Catalytic Functionalization of Nitrogen Compounds: From Selective C–H Amination to Photocatalytic Ammonia Synthesis”
14:40-15:00	**홍완표 (가천대)** “Cycloalkyl Fused Dibenzofuran Derived Green Ir(III) Complexes Possessig High Horizontal Emitting Dipole Orientation Ratios and Color Stability”
15:00-15:30	**Break Time (휴식 및 기념촬영)**
	Session II. 좌장: 김민 (충북대)
15:30-16:00	**제 12 회 젊은 유기화학자상 수상 강연 (이민희, 숙명여대)** “Design and synthesis of activatable fluorescent molecules and their applications”
16:00-16:20	**최이삭 (충북대)** “A Way Towards Sustainable Chemistry in C−H Bond Manipulation”
16:20-16:40	**홍승윤 (서울대)** “Main Group Redox Catalysis For Sustainable C−N Cross-Coupling Reactions”
16:40-17:00	**이호재 (한림대)** “Kinetic Control of Metal-Phenolic Complexation for Film Formation”

12/03 (Sat)

09:00-11:00	**유기분과 발전방안 논의**
11:00-11:10	**폐회식**

박 정 우 (Jung-Woo Park)

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Ph.D. (2012)	Department of Chemistry, Yonsei University (Prof. Chul-Ho Jun)
B.Sc. (2005)	Department of Chemistry, Yonsei University

Position

2022 – present	Assistant Professor, Department of Chemistry, Mokpo National University, Korea
2017 – 2022	Senior Research Fellow (Young Scientist Fellow), Center for Catalytic Hydrocarbon Functionalizations (Director: Prof. Sukbok Chang), IBS, Korea
2014 – 2017	Post-doc., Department of Chemistry, University of California at Irvine, USA (Prof. Vy M. Dong)
2012 – 2013	Post-doc., Department of Chemistry, Yonsei University, Korea (Prof. Chul-Ho Jun)

Representative Publications

1. **Park, J.-W.***, Cobalt-Catalyzed Alkyne Hydrosilylation as a New Frontier to Selectively Access Silyl-Hydrocarbons" *Chem. Commun.* **2022**, *58*, 491-504.

2. Sahoo, M. K.; Kim, D.; Chang, S.; **Park, J.-W.***, Regioselective Access to α-Vinylsilanes and α-Vinylgermanes by Cobalt-catalyzed Migratory Hydrofunctionalization of 2-Alkynes, *ACS Catal.* **2021**, *11*, 12777-12784.

3. Kim, Y. B.; Kim, D.; Dighe, S. U.; Chang, S.; **Park, J.-W.***, Cobalt-Hydride-Catalyzed Hydrosilylation of 3-Alkynes Accompanying π-Bond Migration, *ACS Catal.* **2021**, *11*, 1548-1553.

4. Lee, M.; Jung, H.; Kim, D.; **Park, J.-W.***; Chang, S.* Modular Tuning of Electrophilic Reactivity of Iridium Nitrenoids for the Intermolecular Selective α-Amidation of β-Keto Esters, *J. Am. Chem. Soc.* **2020**, *142*, 11999-12004.

5. **Park, J.-W.***; Kang, B.; Dong, V. M.* Catalytic Alkyne Arylation Using Traceless Directing Group, *Angew. Chem. Int. Ed.* **2018**, *57*, 13598-13602.

Migratory Hydrofunctionalization of Alkynes Enabled by Cobalt Catalysis

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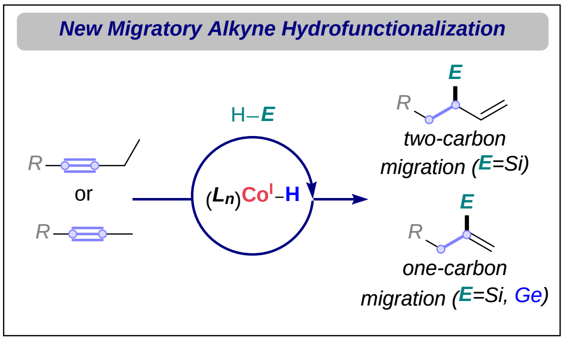
Manipulation of alkynes has been regarded as a substantial synthetic tool to prepare functional olefins, and such strategies based on the direct functional group installation onto the alkyne π-bonds have been generally employed in chemical synthesis. Recently, migratory functionalization of alkynes, where migration of alkyne π-bonds is associated with the introduction of functional groups, has emerged for fabricating synthetically valuable olefins such as allylic compounds.[1] Despite the synthetic potential, previously reported strategies have mostly relied on noble-metal catalysis, and are limited to (i) one-carbon migration and (ii) the generation of allylic functionality. Inspired by the recent endeavors in first-row transition-metal catalysis, our continuous effort has centered on designing base-metal catalyst systems, particularly using cobalt as the model, for the new type of hydrocarbon functionalization. This presentation will show our recent outcomes toward the development of migratory alkyne hydrofunctionalizations enabled by rationally designed cobalt catalysis. By using a cobalt-hydride catalyst bearing a sterically demanding ligand, the selective transformation of internal alkynes such as 3-alkynes and 2-alkynes are successfully accomplished to afford functional alkenes such as branched terminal allylsilanes, α-vinylsilanes, and α-vinylgermanes.[2,3] Mechanistic details of the migratory transformation will also be discussed.



References

[1] For reviews, see: (a) A. M. Haydl, B. Breit, T. Liang, M. J. Krische, *Angew. Chem., Int. Ed.* **2017**, *56*, 11312-11325; (b) P. Koschker, B. Breit, *Acc. Chem. Res.* **2016**, *49*, 1524-1536.

[2] Y. B. Kim, D. Kim, S. U. Dighe, S. Chang, J.-W. Park, *ACS Catal.* **2021**, *11*, 1548-1553.

[3] M. K. Sahoo, D. Kim, S. Chang, J.-W. Park, *ACS Catal.* **2021**, *11*, 12777-12784.

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B.Sc. (2015)	Department of Chemistry, Yonsei University

Position

2022 – present	Assistant Professor, Department of Chemistry, Dong-A University, Korea
2020 – 2022	Post-doc., Department of Chemistry, University of Chicago, USA (Prof. Guangbin Dong)
2020 – 2020	Post-doc., Department of Chemistry, Sungkyunkwan University, Korea (Prof. Jaesook Yun)

Representative Publications

1. **Jang, W. J.**; Woo, J.; Yun, J. “Asymmetric Conjugate Addition of Chiral Secondary Borylalkyl Copper Species” *Angew. Chem., Int. Ed.* **2021**, *60*, 4616.

2. **Jang, W. J.**; Yun, J. “Catalytic Asymmetric Conjugate Addition of a Borylalkyl Copper Complex for Chiral Organoboronate Synthesis” *Angew. Chem., Int. Ed.* **2019**, *58*, 18131.

3. **Jang, W. J.**; Yun, J. “Copper-Catalyzed Tandem Hydrocupration and Diastereo- and Enantioselective Borylalkyl Addition to Aldehydes” *Angew. Chem., Int. Ed.* **2018**, *57*, 12116.

4. **Jang, W. J.**; Song, S. M.; Moon, J. H.; Lee, J. Y.; Yun, J. “Copper-Catalyzed Enantioselective Hydroboration of Unactivated 1,1-Disubstituted Alkenes” *J. Am. Chem. Soc.* **2017**, *139*, 13660.

5. Yoon, W. S.†; **Jang, W. J.†**; Yoon, W.; Yun, H.; Yun, J. “Copper-catalysed asymmetric reductive cross-coupling of prochiral alkenes” *Nat. Commun.* **2022**, *13*, 2570. (**†Co-first authors**)

6. Han, J. T.†; **Jang, W. J.†**; Kim, N.; Yun, J. “Asymmetric Synthesis of Borylalkanes via Copper-Catalyzed Enantioselective Hydroallylation” *J. Am. Chem. Soc.* **2016**, *138*, 15146. (**†Co-first authors**)

Oxidant-induced Direct α-Alkylation of Ketones Enabled by Ir-B Cooperative Catalysis

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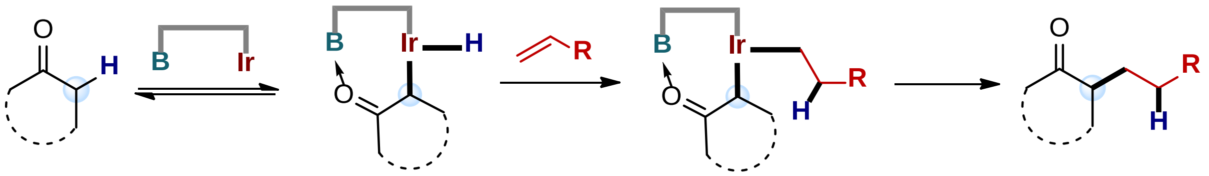
The α-alkylation of carbonyl compounds is a fundamental transformation in the formation of carbon-carbon (C–C) bonds in organic synthesis. Among the various carbonyl functionalization methods, enolate alkylation is one of the most general approaches for introducing carbon substituents at carbonyl α-positions. Although these fundamental organic transformations have been widely utilized in organic synthesis in academic and industrial settings, the reactions have displayed significant environmental and safety concerns from a sustainability viewpoint. Hence, developing more sustainable and efficient reactions would be highly desirable. This presentation will describe a strategy for an oxidant-induced direct α-C–H alkylation of ketones using simple olefins as the alkylating agent enabled by transition metal/Lewis acid cooperative catalysis.[1] In the presence of an Ir/B catalyst, this transformation provides a α-alkylation product in a simple one-step operation. Mechanistic investigations of these reactions, including radical trapping, control experiments, deuterium labeling, and kinetic studies, will be discussed in detail.



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1. Jang, W. J.; Tugwell, T. H.; Choi, S.; Liu, P.; Dong, G. *Manuscript Preparation*.

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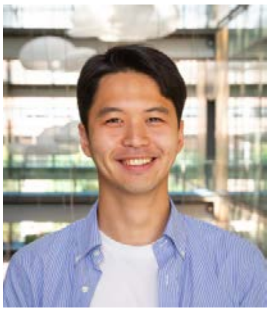

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2019 – 2021	Postdoctoral Research Associate, Department of Chemistry, Princeton University, USA (Prof. Paul J. Chirik)
2019	Research Fellow, Institute for Basic Science, Korea
2017	Visiting Research, University of Utah, USA (Prof. Matthew S. Sigman)

Representative Publications

1. **Park, Y.**; Kim, S.; Tian, L.; Zhong, H.; Scholes, G. D.; Chirik, P. J. Visible Light Enables Catalytic Formation of Weak Chemical Bonds with Molecular Hydrogen. *Nature Chem.* **2021**, *13*, 969-976.

2. **Park, Y.**; Tian, L.; Kim, S.; Pabst, T. P.; Kim, J.; Scholes, G. D.; Chirik, P. J. Visible Light-Driven, Iridium-Catalyzed Hydrogen Atom Transfer: Mechanistic Studies, Identification of Intermediates and Catalyst Improvements. *JACS Au* **2022**, *2*, 407-418.

3. **Park, Y.**; Semproni, S. P.; Zhong, H. Chirik, P. J. Synthesis, Electronic Structure, and Reactivity of a Planar Four-Coordinate, Cobalt-Imido Complex. *Angew. Chem. Int. Ed.* **2021**, *60*, 14376-14380.

4. **Park, Y.**; Chang, S. Asymmetric Formation of γ-Lactams via C–H Amidation Enabled by Chiral Hydrogen-Bond-Donor Catalysts. *Nature Catal.* **2019**, *2*, 219–227.

5. Hong, S. Y.†; **Park, Y.†**; Hwang, Y.; Kim, Y.; Baik, M.-H.; Chang, S. Selective Formation of γ-Lactams via C–H Amidation Enabled by Tailored Iridium Catalysts. *Science* **2018**, *359*, 1016–1021. (†Co-first authors)

6. **Park, Y.**; Heo, J.; Baik, M.-H.; Chang, S. Why is the Ir(III)-Mediated Amido Transfer So Much Faster than the Rh(III)-Mediated Reaction? A Combined Experimental and Computational Study. *J. Am. Chem. Soc.* **2016**, *138*, 14020-14029.

Catalytic Functionalization of Nitrogen Compounds:

From Selective C–H Amination to Photocatalytic Ammonia Synthesis

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Nitrogen-containing molecules are of great importance in the field of organic chemistry, inorganic synthesis, and material science owing to their unique chemical and physical properties.[1] In this talk, I will showcase how a well-defined molecular catalyst could give an efficient and selective access to various classes of nitrogen compounds. In the first part of the talk, mechanistically driven discovery of novel class of amide agents will be discussed. Extensive Investigation on rhodium-catalyzed C–H amidation with organic azide led us to introduce 1,4,2-dioxazol-5-ones as highly efficient amide precursor.[2] An integrated low-temperature NMR kinetics and computational study revealed a key role of high-valent metal-nitrenoid intermediate,[3] and further efforts to utilize its reactivity enabled to design a new array of iridium catalysts for γ-lactam formation.[4] Asymmetric[5] and site-selective[6] catalysis has been also achieved by utilizing non-covalent interactions. The second part of the talk will focus on the development of bifunctional photocatalysts for ammonia synthesis. Proton-coupled electron transfer using molecular hydrogen as a terminal reductant is an attractive strategy for synthesizing weak element-hydrogen bonds, but the intrinsic thermodynamics presents a challenge for reactivity. I will describe the direct photocatalytic synthesis of weak element-hydrogen bonds of unsaturated organic molecules[7] as well as metal-nitrogen compounds.[8] Activation of molecular hydrogen occurs in the ground state and the resulting iridium hydride harvests visible light to enable spontaneous formation of weak chemical bonds near thermodynamic potential without any by-product. Transient absorption spectroscopic studies revealed a triplet-triplet energy transfer as the photophysical initiation process. Identification of catalyst deactivation pathway led to a design of the next-generation catalyst with improved photostability and better catalytic performance.[9]

References

1. Park, Y.; Kim, Y.; Chang S. *Chem. Rev.* **2017**, *117*, 9247.
2. Park, Y.; Park, K. T.; Kim, J. G.; Chang S. *J. Am. Chem. Soc.* **2015**, *137*, 4534.
3. Park, Y.; Heo, J.; Baik, M.-H.; Chang, S. *J. Am. Chem. Soc.* **2016**, *138*, 14020.
4. Hong, S. Y.†; Park, Y.†; Hwang, Y.; Kim, Y. B.; Baik, M.-H.; Chang, S. *Science* **2018**, *359*,1016. († denotes co-first authors)
5. Park, Y.; Chang, S. *Nat. Catal.* **2019**, *2*, 219.
6. Jung, H.; Schrader, M.; Kim, D.; Baik, M.-H.*; Park, Y.*; Chang, S.* *J. Am. Chem. Soc.* **2019**, *141*, 15356.
7. Park, Y.; Kim, S.; Tian, L.; Zhong, H.; Scholes, G. D.; Chirik, P. J. *Nat. Chem.* **2021**, *13*, 969.
8. Park, Y.; Semproni, S. P.; Zhong, H. Chirik, P. J. *Angew. Chem. Int. Ed.* **2021**, *60*, 14376.
9. Park, Y.†; Tian, L.†; Kim, S.; Pabst, T. P.; Kim, J.; Scholes, G. D.; Chirik, P. J. *JACS Au* **2022**, *2*, 407. († denotes co-first authors)

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Education

Ph.D. (2011)	Department of Chemistry, Purdue University (Prof. Pilip L. Fuchs)
B.Sc. (2004)	Department of Industrial Chemistry, Hanyang University

Position

2022 – present	Assistant Professor, Department of Chemistry, Gachon University, Korea
2020 – 2022	Assistant Professor, Department of Advanced Materials and Chemical Engineering, Daegu Catholic University, Korea
2013 – 2020	Associate Researcher, LG Chem
2011 – 2013	Post-doc., Department of Chemistry, University of Wisconsin, USA (Prof. Shannon S. Stahl)

Representative Publications

1. Kim, S. C., Hong, W. P.*, Lee, J. Y.* “Cycloalkyl Fused Dibenzofuran Derived Green Ir(III) Complexes Possessig High Horizontal Emitting Dipole Orientation Ratios and Color Stability at High Doping Concentrations” *Adv. Opt. Mater.* **2022**, ASAP.

2. Tagare, J.*, Hong, W. P.* “Recent Progress in Imidazole Based Efficient near Ultraviolet/Blue Hybridized Local Charge Transfer (HLCT) Characteristics Fluorophores for Organic Light-Emitting Diodes” *J. Mater. Chem. C.* **2022**, ASAP.

2. Shin, I.; Lim, H. N.*, Hong, W. P.* “1,4-Azaborines: Origin, Modern Synthesis, and Applications as Optoelectronic Materials” *Synthesis* **2022**, *54*, 570.

3. Kim, D. S., Yoon, S. J., Lee, K. H., Lee, J. Y.*, Hong, W. P.* “Starburst Type Benzofuroindolocarbazole Donor for High Efficiency and Long Lifetime in Thermally Activated Delayed Fluorescence Emitters” *Adv. Opt. Mater.* **2020**, *9*, 2001432.

Cycloalkyl Fused Dibenzofuran Derived Green Ir(III) Complexes Possessig High Horizontal Emitting Dipole Orientation Ratios and Color Stability

Wan Pyo Hong

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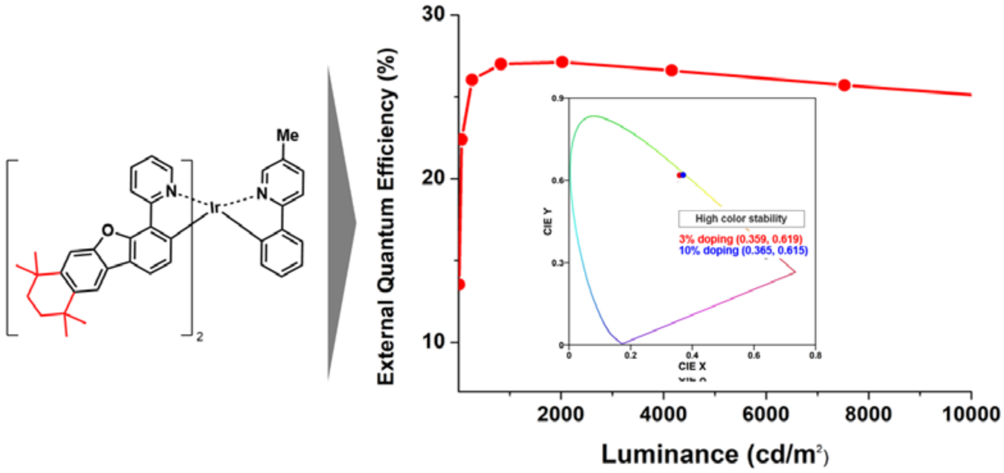
Last three decades tremendous progress have been made in the important filed of Organic Light Emitting Diodes (OLEDs). Over such a long journey, the comprehensive performance of the phosphorescent materials have been greatly improved in terms of color purity, luminous efficiency and the operational lifetime.

As an advancement to this, we uncovered the photophysical characteristics and electroluminescence of three green Ir(III) complexes with newly created cycloalkyl fused dibenzofuran ligands. The finding demonstrate that the cycloalkyl fused dibenzofuran ligand can modify the intermolecular interactions and alter the horizontal emitting dipole orientation ratios of the dopants in phosphorescent organic light-emitting diodes (PhOLEDs).

Among the three dopants, Ir(TBF)2(mppy) exhibited a high photoluminescence quantum yield of 0.96 and a high horizontal emitting dipole orientation ratio of 0.80. Further, an optimized PhOLED using Ir(TBF)2(mppy) demonstrated a peak external quantum efficiency (EQE) of 27.1% and a very small efficiency roll-off with a high EQE of 25.1% at 10000 cd m−2.



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Education

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M.S. (2008)	Department of Chemistry, Dankook University (Prof. Jong Seung Kim)
B.Sc. (2006)	Department of Chemistry, Dankook University

Position

2020 – present	Associate Professor, Department of Chemistry, Sookmyung Women's University, Korea
2015 – 2020	Assistant Professor, Department of Chemistry, Sookmyung Women's University, Korea
2012 – 2015	Post-doc., Department of Chemistry, The University of Texas at Austin, Texas, USA (Prof. Jonathan L. Sessler)

Representative Publications

1. S. Y. Park, K. Kim, D.-H. Cho, E.-Y. Jo, C. Kang, **M. H. Lee**, 'Indole-based fluorescent chemosensor targeting to autophagosome', *Chem. Commun.*, **2022**, *58*, 2886-2889.

2. S. A Yoon, M. Maiti, Y. Cha, K. K. Athul, S. Bhuniya, **M. H. Lee**, 'Cell-specific activation of gemcitabine by endogenous H_2S stimulation and tracking through simultaneous fluorescence turn-on', *Chem. Commun.*, **2021**, *57*, 9614.

3. S. J. Kim, S. Y. Park, S. A. Yoon, C. Kim, C. Kang, **M. H. Lee**, 'Naphthalimide-4-(4-nitrophenyl)thiosemicarbazide: A fluorescent probe for simultaneous monitoring of viscosity and nitric oxide in living cells', *Anal. Chem.* **2021**, *93*, 4391-4397. (Supplementary cover article)

4. S. A. Yoon, J. Chun, C. Kang, **M. H. Lee**, 'Self-Calibrating Bipartite Fluorescent Sensor for Nitroreductase Activity and Its Application to Cancer and Hypoxic Cells', *ACS Applied Bio Materials*, **2021**, *4*, 2052-2057. (Supplementary cover article)

5. S. Y. Park, S. A. Yoon, Y. Cha, **M. H. Lee**, 'Recent advances in fluorescent probes for cellular antioxidants: Detection of NADH, hNQO1, H_2S, and other redox biomolecules', *Coord. Chem. Rev.*, **2020**, *428*, 213613.

6. J. H. Joo, K. Kim, S. Y. Park, J. Lee, C. Kang, **M. H. Lee,** 'Fluorogenic sensing of mitochondrial electron transferring dehydrogenase using a trimethyl lock quinone-linked hemicyanine', *Sensors and Actuators B: Chemical*, **2020**, *322*, 128578.

7. J. H. Joo, M. Won, S. Y. Park, K. Park, D. S. Shin, J. S. Kim, **M. H. Lee,** 'A dicyanocoumarin-fused quinolinium based probe for NAD(P)H and its use for detecting glycolysis and hypoxia in living cells and tumor spheroids', *Sensors and Actuators B: Chemical*, **2020**, *320*, 128360.

Design and synthesis of activatable fluorescent molecules and their applications

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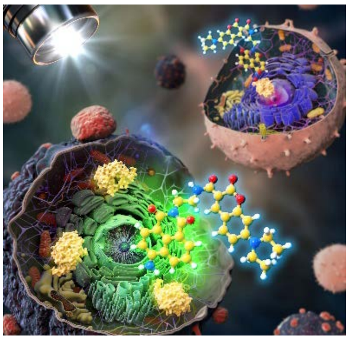
Quantitative determination of specific analytes is essential for a variety of applications ranging from life sciences to environmental monitoring. Optical sensing allows for non-invasive measurements within biological milieus, parallel monitoring of multiple samples, and less invasive imaging. Among the optical sensing methods currently being explored, small molecule-based fluorescence sensing has received particular attention as a technique with the potential to provide precise and quantitative analyses and real-time fluorescence monitoring. So far, our laboratory has developed a variety of sensing probes using activatable fluorescent molecules for sensing, imaging, and biomedical applications. Today, I will talk about development of activatable fluorescent molecules capable of sensing several bioactive components such as hydrogen sulfide (H_2S), human NAD(P)H:quinone oxidoreductase 1 (hNQO1), nitroreductase (NTR), reduced nicotinamide adenine dinucleotide (NADH), and exploring dual sensing of nitric oxide (NO) and cellular viscosity, recently reported in our laboratory. The basic design concepts involving small fluorescent molecules composed of a fluorescent reporter and recognition moiety that undergo a fluorogenic reaction in response to analytes will be described. In addition, some biological results demonstrating the validation of fluorescent molecules' operation will be presented in a variety of biological models such as live cancer cells, cancer cell spheroids and tissues.



References

1. S. A Yoon, M. Maiti, Y. Cha, K. K. Athul, S. Bhuniya, **M. H. Lee**, 'Cell-specific activation of gemcitabine by endogenous H_2S stimulation and tracking through simultaneous fluorescence turn-on', *Chem. Commun.*, **2021**, *57*, 9614.
2. S. J. Kim, S. Y. Park, S. A. Yoon, C. Kim, C. Kang, **M. H. Lee**, 'Naphthalimide-4-(4-nitrophenyl)thiosemicarbazide: A fluorescent probe for simultaneous monitoring of viscosity and nitric oxide in living cells', *Anal. Chem.* **2021**, *93*, 4391-4397. (Supplementary COVER Article)
3. S. A. Yoon, J. Chun, C. Kang, **M. H. Lee**, 'Self-Calibrating Bipartite Fluorescent Sensor for Nitroreductase Activity and Its Application to Cancer and Hypoxic Cells', *ACS Applied Bio Materials*, **2021**, *4*, 2052-2057. (Supplementary cover article)
4. S. Y. Park, S. A. Yoon, Y. Cha, **M. H. Lee**, 'Recent advances in fluorescent probes for cellular antioxidants: Detection of NADH, hNQO1, H_2S, and other redox biomolecules', *Coord. Chem. Rev.*, **2020**, *428*, 213613.

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Position

2022 – present Assistant Professor, Department of Chemistry, Chungbuk National University, Korea
2021 – 2022 Research Associate, University of Wisconsin-Madison, USA (Prof. Jennifer M. Schomaker)

Representative Publications

1. Choi, I.; Trenerry, M. J.; Lee, K. S.; King, N.; Berry, J. F.; Schomaker J. M. Divergent C–H Amidations and Imidations by Tuning Electrochemical Reaction Potentials *ChemSusChem*, Accepted

2. Choi, I.; Hou, X.; Ackermann, L. A Strategy for Site- and Chemoselective C−H Alkenylation through Osmaelectrooxidative Catalysis *Angew. Chem. Int. Ed.* **2021**, *60*, 27005–27012.

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A Way Towards Sustainable Chemistry in C−H Bond Manipulation

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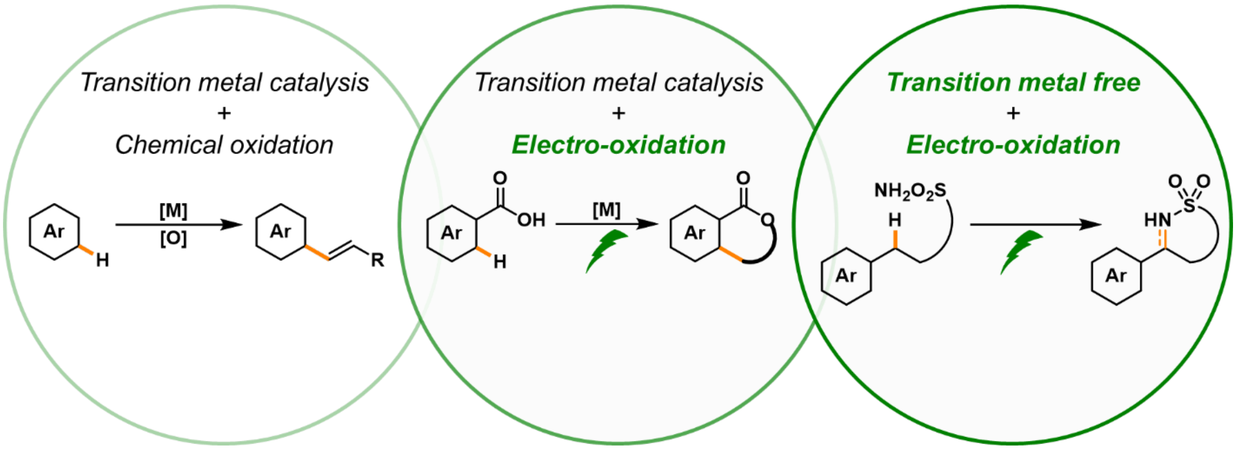
C–H bond functionalization provides chemists with efficient and step-economical strategies in organic syntheses. However, oxidative/reductive transformations still require suprastoichiometric amounts of terminal oxidants/reductants, which generate a large quantity of chemical wastes and thereby prevents sustainable chemistry.



In this talk, I present my recent research efforts toward the establishment of sustainable organic syntheses, spanning from transition metal-catalyzed oxidative C–H activation[1] to metallaelectrooxidative annulation[2] and electro-tunable divergent C–H functionalization.[3]

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2020 – 2021	Research Fellow, Institute for Basic Science (IBS), Korea (Prof. Sukbok Chang)

Representative Publications

1. **S. Y. Hong** and A. T. Radosevich*, Chemoselective Primary Amination of Aryl Boronic Acids by $P^{III}/P^{V}=O$ Catalysis: Synthetic Capture of the Transient Nef Intermediate HNO, *J. Am. Chem. Soc.*, **2022**, *144*, 8902–8907.
2. **S. Y. Hong**, Y. Hwang, M. Lee, and S. Chang*, Mechanism-Guided Development of Transition-Metal Catalyzed C–N Bond-Forming Reactions Using Dioxazolones as the Versatile Amidating Source, *Acc. Chem. Res.*, **2021**, 54, 2683–2700.
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Main Group Redox Catalysis
For Sustainable C–N Cross-Coupling Reactions

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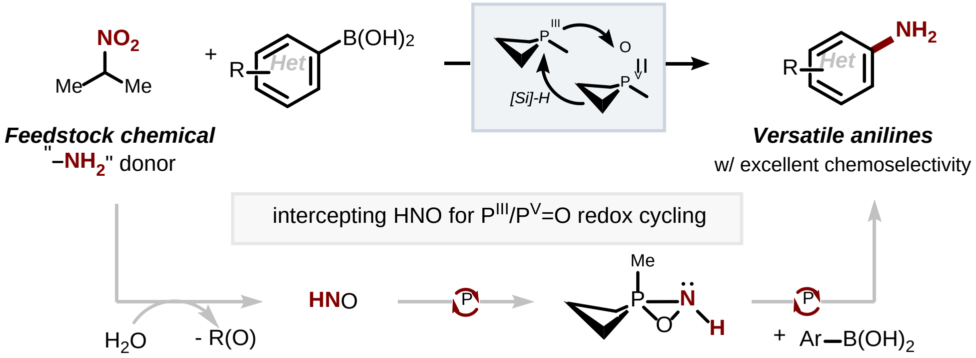
Catalytic reactions that construct carbon-nitrogen (C−N) bonds are of sustained interest in the synthetic organic community since they can offer a streamlined route to useful nitrogen-containing motifs commonly encountered in natural products and clinical drugs. The prevailing strategy to meet such demands in industrial settings is currently shaped by transition metal-catalyzed methods, as exemplified by Buchwald-Hartwig couplings. Despite the significant strides in this area, increasing emphasis has been placed on developing a cost-effective and sustainable alternative that omits a transition metal and repositions the main group elements from the catalytic periphery to the center of bond making and breaking during catalysis.

In this talk, a functionally and mechanistically distinct main group-catalyzed method for the chemoselective formation of primary amines will be presented. Key to success is accentuating the biphilic reactivity of small-ring phosphacycles that enables the use of nitroalkanes—many of which are stable and inexpensive—as amination reagents for C−N coupling chemistry.[1] Specifically, a phosphetane-based catalyst operating within $P^{III}/P^{V}{=}O$ redox cycling is shown to capture HNO, generated *in situ* by Nef decomposition of readily available 2-nitropropane, to install the primary amino group at aryl C_{sp2} centers selectively.[2] This method represents a sustainable way of preparing versatile nitrogen-containing products that not only shows complementarity in scope and selectivity to existing methods but bypasses commonly faced challenges (e.g. overamination and metal contamination) in transition metal catalysis.



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Representative Publications

1. Lee, H., Park, J., Kim, N., Youn, W., Yun, G., Han, S. Y., Nguyen, D. T. and Choi, I. S.* “Cell-in-Catalytic-Shell Nanoarchitectonics: Catalytic Empowerment of Individual Living Cells by Single-Cell Nanoencapsulation” *Adv. Mater.* **2022**, *34*, 2201247.

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Kinetic Control of Metal-Phenolic Complexation for Film Formation

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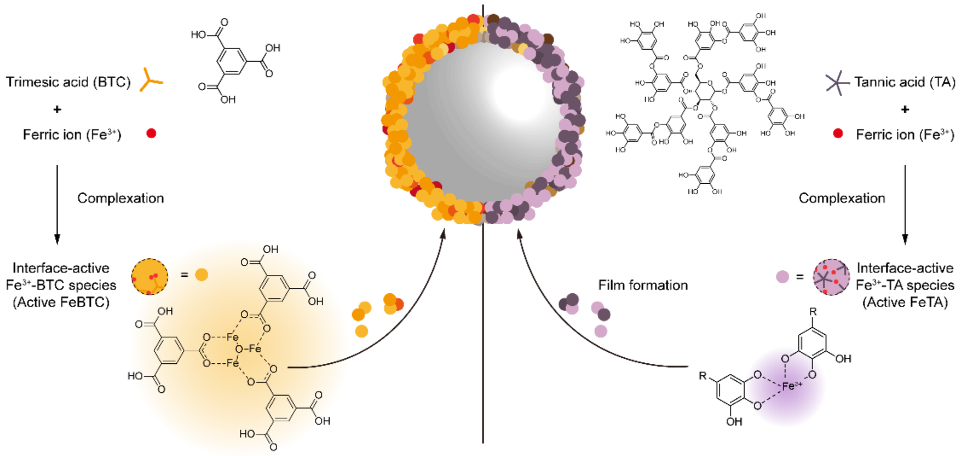
Metal-phenolic complex, representatively from supramolecular self-assembly of Fe^{3+} and tannic acid (TA), have widely been studied for understanding bio-inspired adhesion and functional coatings of various materials, including battery separators and living cells. However, the mechanism of Fe^{3+}-TA film formation had remained elusive, and it was generally accepted that molecules containing catechol (1,2-dihydroxybenzene) or pyrogallol (1,2,3-trihydoxybenzene) moieties could make a molecular adhesiveness. Recently, the iron gall ink (IGI)-based method provided an inkling that the film growth depended on the reaction rate of supramolecular complexation. For example, using Fe^{2+} instead of Fe^{3+} led slow but continuous Fe^{3+}-TA film formation by providing Fe^{3+} ion through the steady air-oxidation of Fe^{2+} to Fe^{3+} in solution. A more tightly controlled kinetic study provided the first-level understanding of the film formation mechanism. In addition, an organic ligand, beyond the phenolics, that generates molecular adhesiveness has been proposed, allowing us to obtain the chemical intuition of the adhesive coating. In this session, a summary of our recent efforts to design adhesive molecules and to elucidate their working mechanisms will be given.



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