

Appendices

1. Khima Pandey, **Pinku Kaswan**, Saroj, and Anil Kumar, "Synthesis of 2-carbonylimidazo[1,2-*a*]pyridines *via* iodine-mediated intramolecular cyclization of 2-amino-*N*-propargylpyridinium bromides" *Chemistry Select* **2016**, 1, 6669–6672.
2. **Pinku Kaswan**, Ganesh M. Shelke, V. Kameswara Rao, and Anil Kumar, "Hydroxy-group-facilitated vinylic iodination of *ortho*-vinylnaphthols using molecular iodine" *Synlett* **2016**, 27, 2553–2556.
3. **Pinku Kaswan**, Nitesh Kumar Nandwana, Brenton DeBoef, and Anil Kumar, "Vanadyl acetylacetonate catalyzed methylenation of imidazo[1,2-*a*]pyridines by using dimethylacetamide as a methylene source: Direct access to bis(imidazo[1,2-*a*]pyridin-3-yl)methanes" *Advanced Synthesis & Catalysis* **2016**, 358, 2108–2115
4. **Pinku Kaswan**, Ashley Porter, Kasiviswanadharaju Pericherla, Marissa Simone, Sean Peters, Anil Kumar, and Brenton DeBoef, "Oxidative cross-coupling of sp^3 - and sp^2 -hybridized C–H bonds: Vanadium-catalyzed aminomethylation of imidazo[1,2-*a*]pyridines" *Organic Letters* **2015**, 17, 5208–5211.
5. Hitesh K Saini, **Pinku Kaswan**, Kasiviswanadharaju Pericherla, and Anil Kumar "Synthesis of naphtho-fused imidazo[1,2-*a*]pyridines *via* copper catalyzed cascade reactions" *Asian Journal of Organic Chemistry* **2015**, 4, 1380–1385.
6. **Pinku Kaswan**, V. Kameswara Rao, Keykavous Parang and Anil Kumar, "Indium triflate catalyzed microwave-assisted alkenylation of methoxyphenols: Synthesis of indenenes and chromenes" *Organic and Biomolecular Chemistry* **2015**, 13, 11072–11077.
7. **Pinku Kaswan**, V. Kameswara Rao, Ganesh M. Shelke, Ashley Ryan, Mukund Jha, Anil Kumar, "Iodine-mediated, microwave-assisted synthesis of 1-arylnaphthofurans *via* cyclization of 1-(1'-arylviny)-2-naphthols" *Synthesis* **2015**, 47, 3990–3996.
8. Kasiviswanadharaju Pericherla, **Pinku Kaswan**, Khima Pandey, and Anil Kumar, "Recent developments in the synthesis of imidazo[1,2-*a*]pyridines" *Synthesis* **2015**, 47, 887–912.
9. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, Deepshikha Purohit, and Anil Kumar, "Synthesis of 5,7-diarylpyrazolo[1,5-*a*]pyrimidines *via* KOH mediated tandem reaction of 1*H*-pyrazol-3-amines and chalcones" *Tetrahedron Letters* **2015**, 56, 549–553.
10. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, Hitesh Kumar Saini and Anil Kumar, "One-pot, three component tandem reaction of 2-aminopyridines, acetophenones and

aldehydes: Synthesis of 3-arylimidazo[1,2-*a*]pyridines” *RSC Advances* **2015**, 5, 3670–3677.

11. Nitesh Kumar Nandwana, Kasiviswanadharaju Pericherla, **Pinku Kaswan** and Anil Kumar, “Synthesis of novelazole-fused quinazolines *via* one-pot, sequential Ullmann-type coupling and intramolecular dehydrogenative C–N bonding” *Organic and Biomolecular Chemistry* **2015**, 13, 2947–2950.
12. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, Rajnikant, Anil Kumar, “Synthesis of 3-arylimidazo[1,2-*a*]pyridines *via* CuCl₂ catalyzed tandem dual carbon-nitrogen bonding” *Tetrahedron* **2014**, 70, 8539–8544.
13. Kasiviswanadharaju Pericherla, **Pinku Kaswan**, Poonam Khedar, Bharti Khungar, Keykavous Parang and Anil Kumar, “Copper catalyzed tandem oxidative C–H amination/cyclizations: Direct access to imidazo[1,2-*a*]pyridines” *RSC Advances* **2013**, 3, 18923–18930.
14. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, Anil Kumar, “Ligand-free, copper-catalyzed ullmann-type C–N coupling: Regioselective synthesis ofazole-substituted imidazo[1,2-*a*]pyridines” *Synlett* **2013**, 24, 2751–2757.

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■ Organic & Supramolecular Chemistry

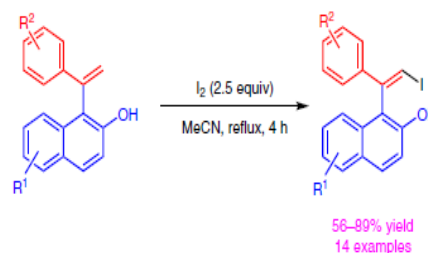
Synthesis of 2-Carbonylimidazo[1,2-*a*]pyridines via Iodine-mediated Intramolecular Cyclization of 2-Amino-*N*-propargylpyridinium BromidesKhima Pandey, Pinku Kaswan, Saroj, and Anil Kumar^{*[a]}

A facile and novel, metal free method has been described for the synthesis of 2-carbonylimidazo[1,2-*a*]pyridines by iodine mediated intramolecular cyclization of 2-amino-*N*-propargylpyridinium bromides in the presence of a base. Various

substituted imidazo[1,2-*a*]pyridine derivatives were obtained in good to excellent yields (45-89%) and good functional group tolerance was observed.

Synlett 2016, 27, 2553–2556**Synlett**

P. Kaswan et al.

Letter**Hydroxy-Group-Facilitated Vinylic Iodination of *ortho*-Vinylnaphthols Using Molecular Iodine**Pinku Kaswan
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Vanadyl Acetylacetonate Catalyzed Methylenation of Imidazo[1,2-*a*]pyridines by Using Dimethylacetamide as a Methylene Source: Direct Access to Bis(imidazo[1,2-*a*]pyridin-3-yl)methanes

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Abstract: An efficient protocol has been developed for the methylenation of imidazo[1,2-*a*]pyridines using dimethylacetamide (DMA) as methylene source in the presence of vanadyl acetylacetonate [VO(acac)₂] as the catalyst and iodobenzene diacetate as the oxidant. The reaction involves coupling of sp³- and sp²-hybridized carbons and proceeds through the formation of an iminium ion. A wide va-

riety of imidazo[1,2-*a*]pyridines were converted to bis(imidazo[1,2-*a*]pyridin-3-yl)methanes in good to excellent yields. A gram-scale reaction demonstrated the potential for the scale-up processes.

Keywords: bis(imidazo[1,2-*a*]pyridin-3-yl)methanes; C–H functionalization; imidazo[1,2-*a*]pyridines; methylenation; vanadium catalyst

Organic Letters 2015, 17, 5208–5211

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Oxidative Cross-Coupling of sp³- and sp²-Hybridized C–H Bonds: Vanadium-Catalyzed Aminomethylation of Imidazo[1,2-*a*]pyridines

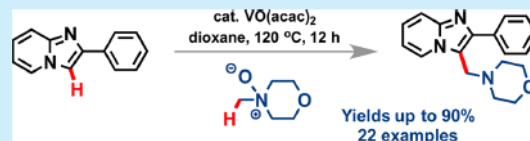
Pinku Kaswan,^{†,§} Ashley Porter,^{‡,§} Kasiviswanadharaju Pericherla,^{†,‡} Marissa Simone,^{‡,‡} Sean Peters,[‡] Anil Kumar,^{*,†} and Brenton DeBoef^{*,‡}

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Supporting Information

ABSTRACT: The vanadium-catalyzed oxidative coupling of substituted 2-arylimidazo[1,2-*a*]pyridines to *N*-methylmorpholine oxide, which acts as both a coupling partner and an oxidant, has been achieved. This reaction was applied to various substituted imidazo[1,2-*a*]pyridine and indole substrates, resulting in yields as high as 90%. Mechanistic investigations indicate that the reaction may proceed via a Mannich-type process. This work demonstrates how oxidative aminomethylation can be used as a useful method to introduce tertiary amines into heterocycles, thus providing an alternative method for conventional Mannich-type reactions.



Synthesis of Naphtho-Fused Imidazo[1,2-*a*]pyridines via Copper Catalyzed Cascade Reactions

Hitesh Kumar Saini, Pinku Kaswan, Kasiviswanadharaju Pericherla and Anil Kumar*

Dedicated to Prof. S. M. S. Chauhan on his 64th birthday

Abstract: A highly efficient copper catalyzed one-pot tandem protocol have been developed for the synthesis of novel naphtho-fused imidazo[1,2-*a*]pyridines. The transformation involves Knoevenagel condensation followed by chemoselective cross-coupling reaction via carbon–carbon bond cleavage. The protocol could tolerate a variety of functional groups and provided naphtho[1',2':4,5]imidazo[1,2-*a*]pyridines in moderate to excellent (35–91%) yields. The photophysical studies of naphtho-fused imidazo[1,2-*a*]pyridines showed bathochromic shift and they emitted with high fluorescence quantum yields.

synthesized using Rh^[8] and Pd-catalyzed^[9] reactions. But there is much interest in the development of Cu-catalyzed cascade reactions for the synthesis of structurally diversified π -expanded heterocycles in one pot because of the convenience, low cost, and high efficiency.^[10]

With the advent of interest in π -expanded heterocyclic compounds, we envisioned that a simple and straightforward synthesis of naphtho-fused imidazo[1,2-*a*]pyridines can offer new opportunities to develop materials for future optoelectronic applications. We, therefore directed our efforts to develop an efficient and convenient synthetic protocol for naphtho-fused



Indium triflate catalyzed microwave-assisted alkenylation of methoxyphenols: synthesis of indenenes and chromenes†‡

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In(OTf)₃ catalyzed microwave-assisted alkenylation of methoxyphenols was investigated. Exclusive formation of either indenenes or chromenes was observed depending on the position of the methoxy group on phenol. The structures of 1*H*-inden-4-ol derivatives (**4a–e**) and 4*H*-chromene derivatives (**5a–j**) were established by NMR (¹H & ¹³C) and high-resolution mass spectra, which were further supported by single crystal X-ray analysis of **4c** and **5a**.

Introduction

Alkenylation or hydroarylation is a useful reaction for aromatic C–H bond functionalization that leads to the formation of multi-substituted olefins. This reaction has attracted significant attention in recent years due to its high atom- and step-economy. Several catalytic systems based on Lewis acids, salts,

6-hydroxyindenenes have remained sparse. Hence, mild and efficient methods for the construction of indene ring frameworks from commercially available substrates are desirable.

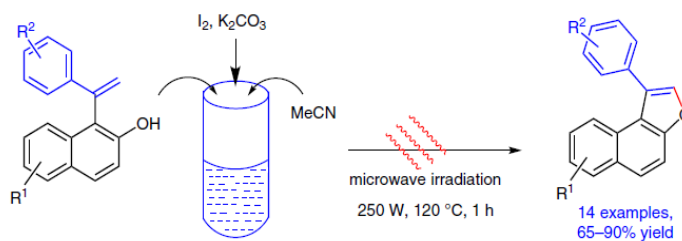
4*H*-Chromenes have also been the subject of consistent interest due to their structural diversity and a wide spectrum of biological activities.^{27–29} Chromene derivatives have also been shown to exhibit photochromism.³⁰ Due to the broad

Iodine-Mediated, Microwave-Assisted Synthesis of 1-Arylnaphthofurans via Cyclization of 1-(1'-Arylvinyloxy)-2-naphthols

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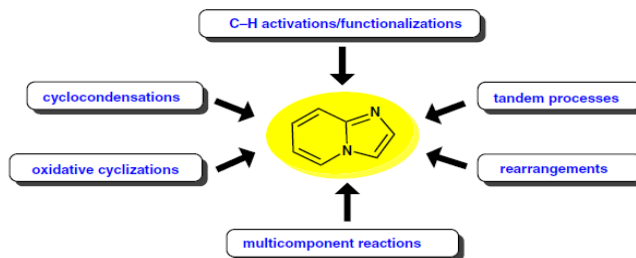
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Recent Developments in the Synthesis of Imidazo[1,2-*a*]pyridines

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Abstract Advances in the last decade for the synthesis of the imidazo[1,2-*a*]pyridine scaffold from various substrates employing approaches such as multicomponent reactions, tandem processes, rearrangement reactions, inter- and intramolecular oxidative/reductive cyclizations, and transition-metal-catalyzed C–H activation are summarized in this review. The mechanisms for the selected transformations are also discussed.

1 Introduction

Among nitrogen-fused azoles, imidazo[1,2-*a*]pyridines have a lead role in the literature because of their wide variety of applications in various disciplines like medicinal chemistry, organometallics, and material science.¹ Although imidazo[1,2-*a*]pyridines are structurally different from benzodiazepines, their pharmacological properties are quite similar to that of benzodiazepine drugs, hence they are termed as nonbenzodiazepines. Molecules with



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Synthesis of 5,7-diarylpyrazolo[1,5-*a*]pyrimidines via KOH mediated tandem reaction of 1*H*-pyrazol-3-amines and chalcones



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ABSTRACT

An efficient and facile protocol has been developed for the synthesis of pyrazolo[1,5-*a*]pyrimidines through the tandem reaction of 3-aminopyrazoles and chalcones in the presence of catalytic amounts of KOH. The reported method offers access to 5,7-diarylpyrazolo[1,5-*a*]pyrimidines in good to excellent yields. A gram-scale reaction has been performed to demonstrate the potency of optimized procedure for the scale-up processes.

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One-pot, three component tandem reaction of 2-aminopyridines, acetophenones and aldehydes: synthesis of 3-arylimidazo[1,2-*a*]pyridines[†]

Pinku Kaswan, Kasiviswanadharaju Pericherla, Hitesh Kumar Saini and Anil Kumar*

A facile synthesis of 3-arylimidazo[1,2-*a*]pyridine derivatives has been achieved through the one-pot, three-component tandem reaction of acetophenones, arylaldehydes and 2-aminopyridines in the presence of a catalytic amount of copper(II) chloride and air as the sole oxidant. The developed one-pot method is atom-economical and utilizes readily available precursors to offer highly functionalized N-fused imidazoles in moderate to good yields (26–82%). The presented tandem process is expected to proceed *via* crossed aldol condensation, Michael addition, copper catalyzed oxidative cyclization and subsequent aromatization.

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Synthesis of novelazole-fused quinazolines *via* one-pot, sequential Ullmann-type coupling and intramolecular dehydrogenative C–N bonding†

Nitesh Kumar Nandwana, Kasiviswanadharaju Pericherla, Pinku Kaswan and Anil Kumar*

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An efficient one-pot sequential procedure is described for the synthesis of novelazole-fused quinazolines through Pd/Cu co-catalyzed, Ullmann-type coupling followed by cross dehydrogenative coupling of various azoles such as 1*H*-imidazole, 1*H*-benzimidazole and 1*H*-1,2,4-triazole with 2-(2-bromophenyl)-1*H*-imidazole/benzimidazoles. The developed strategy has offered good yields (52–81%) of diverse *N*-fused tetra-, penta- and hexa-cyclic frameworks in a single step.

Tetrahedron 2014, 70, 8539–8544



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Synthesis of 3-arylimidazo[1,2-*a*]pyridines *via* CuCl₂ catalyzed tandem dual carbon–nitrogen bonding



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ABSTRACT

A novel tandem approach has been demonstrated for the direct synthesis of bioactive 3-arylimidazo[1,2-*a*]pyridines from chalcones and 2-aminopyridines. The reported tandem reaction is atom-economical and expected to proceed via 1,4-Michael addition followed by copper catalyzed oxidative C–N bonding. Catalytic amount of copper was found to be crucial for the success of tandem reaction and it altered the reaction pathway to furnish entirely new products. This protocol proved to be convenient as reaction proceeds smoothly without the necessity of any ligand in the presence of air as oxidant.

Copper catalyzed tandem oxidative C–H amination/ cyclizations: Direct access to imidazo[1,2-*a*]pyridines†

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A simple and convenient strategy is described for the synthesis of imidazo[1,2-*a*]pyridines *via* inexpensive copper-catalyzed tandem imine formation and intramolecular aerobic oxidative C–H bond amination/ cyclizations. An array of imidazo[1,2-*a*]pyridines were prepared by the reaction of readily available acetophenones and 2-aminopyridines in good to excellent yields (48–92%). The scope of the method was validated by a single step synthesis of Zolimidine, a drug used for peptic ulcers, in 61% yield.

Synlett 2013, 24, 2751–2757

Synlett

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Letter

Ligand-Free, Copper-Catalyzed Ullmann-Type C–N Coupling: Regioselective Synthesis of Azole-Substituted Imidazo[1,2-*a*]pyridines

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Abstract: A simple and highly efficient protocol for the regioselective synthesis of azole-substituted imidazo[1,2-*a*]pyridines has been developed using a ligand-free, copper-catalyzed Ullmann-type C–N coupling of 2-(2-bromophenyl)imidazo[1,2-*a*]pyridines with different azoles and *in situ* generated 1,2,3-triazoles. The reactions proceeded smoothly to furnish azolo-imidazo[1,2-*a*]pyridines in good to excellent yields (65–96%).

Key words: copper, ligand-free C–N coupling, imidazo[1,2-*a*]pyridines, 1,2,3-triazole, CuAAC, tandem reaction

Copper-catalyzed Ullmann-type couplings are traditional methods for forming carbon–heteroatom bonds.¹ However,

alpidem, necopidem and saripidem (anxiolytics), zolimidine (treatment of peptic ulcers), olprinone (cardio-tonic agent), and miroprofen (analgesic).

Although some ligand-promoted C–N couplings have been reported on imidazo[1,2-*a*]pyridines, *ortho*-directed aminations of these privileged motifs are poorly studied.⁵ In a continuation of our efforts towards designing novel heterocycles containing imidazo[1,2-*a*]pyridines,¹⁶ we wish to report a regioselective, ligand-free, copper-catalyzed C–N coupling for the synthesis of azole-substituted imidazo[1,2-*a*]pyridines by the reaction of 2-(2-bromophenyl)imidazo[1,2-*a*]pyridines with different azoles and *in situ* generated 1,2,3-triazoles (Scheme 1).

Oral Presentation

1. **Pinku Kaswan** and Anil Kumar “Synthesis of 3-arylimidazo[1,2-*a*]pyridine, and functionalization of imidazo[1,2-*a*]pyridines using vanadium catalyst” at 23rd ISCB International Conference (ISCBC-2017), SRM University, Chennai, India, February 8-10, 2017.
2. **Pinku Kaswan** and Anil Kumar, “Oxidative cross-coupling of sp³- and sp²-hybridized C–H bonds: Vanadium-catalyzed aminomethylation of imidazo[1,2-*a*]pyridines” at International Conference on Recent Advances in Chemical Sciences (RACS-2015), JECRC University, Jaipur, India, December 28-30, 2015.
3. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, and Anil Kumar, “Copper catalyzed tandem synthesis of 3-arylimidazo[1,2-*a*]pyridines *via* C-N bonding” at National Conference on Frontiers at the Chemistry-Allied Sciences Interface (FCASI-2015), University of Rajasthan, Jaipur, India, March 13-14, 2015.

Poster Presentation

1. **Pinku Kaswan**, Vikki N. Shinde, Anil Kumar, “A sequential Suzuki cross-coupling/direct arylation of 2-arylimidazo[1,2-*a*]pyridine: One-pot access to multi-substituted imidazo[1,2-*a*]pyridine” at National Conference on Organic Chemistry in Sustainable Development: Recent Advances and Future Challenges (OSCD-2015), BITS Pilani, Pilani Campous, India, August 29-30, 2016.
2. **Pinku Kaswan**, Nitesh Kumar Nandwana, Anil Kumar “Synthesis of bis(2-arylimidazo[1,2-*a*]pyridin-3-yl)methane *via* VO(acac)₂ catalyzed reaction of imidazo[1,2-*a*]pyridine with diethylacetamide” at 22nd ISCB International Conference (ISCBC-2016), Department of Chemistry, Uka Tarsadia University, Surat, India, February 6-8, 2016.
3. **Pinku Kaswan**, Nitesh Kumar Nandwana, Anil Kumar, “Synthesis of bis(2-arylimidazo[1,2-*a*]pyridin-3-yl)methane by VO(acac)₂ catalyzed reaction of imidazo[1,2-*a*]pyridine with dimethylacetamide” at International Conference on Nascent Developments in Chemical Sciences: Opportunities for Academia-Industry Collaboration

(NDCS-2015), Department of Chemistry, BITS Pilani, Pilani Campus, India, October 16-18, 2015 (**Best Poster, RSC Award**)

4. **Pinku Kaswan**, Kasiviswanadharaju Pericherla and Anil Kumar, “Tandem three-component reaction of methyl ketones, aldehydes and 2-aminopyridines for rapid synthesis of 3-arylimidazo[1,2-*a*]pyridines in one-pot” at International Symposium On Recent Advances In Medicinal Chemistry (ISRAM-2014) Department of Medicinal Chemistry, (NIPER). S.A.S. Nagar, Punjab, India, September 8-10, 2014.
5. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, Poonam Khedar and Anil Kumar “Copper catalyzed tandem oxidative C–H amination/oxidative cyclizations: Direct access to imidazo[1,2-*a*]pyridines” at 16th CRSI National Symposium in Chemistry, Department of Chemistry, IIT Bombay, Mumbai, February 7-8, 2014.
6. **Pinku Kaswan**, Kasiviswanadharaju Pericherla, and Anil Kumar “Copper catalyzed tandem oxidative C–H amination/oxidative cyclizations: Direct access to imidazo[1,2-*a*]pyridines” at National Conference on Advanced Scientific Development in Chemical Sciences (ASDCS-2014), Department of Chemistry, Deenbandhu Chhotu Ram University of Science & Technology, Murthal, India, March 14, 2014.
7. **Pinku Kaswan**, Kasiviswanadharaju Pericherla and Anil Kumar, “Ligand free CuI-catalyzed amination of 2-(2-bromophenyl)*H*-imidazo[1,2-*a*]pyridines” at 19th ISCB International Conference (ISCBC-2013), Mohanlal Sukhadia University, Udaipur , Rajasthan, March 2-5, 2013.

Brief Biography of the Candidate

[A-4]

Pinku Kaswan belongs to a small village Gagarwas in district Churu of Rajasthan, India. She earned her B.Sc. in 2009 from University of Rajasthan, Jaipur and then she completed her M.Sc. in chemistry from the Department of Chemistry, University of Rajasthan, Jaipur (India) in 2012. In December, 2011 she cleared the Joint CSIR-UGC Test for Junior Research Fellowship and Eligibility for Lectureship (NET) with 60th rank in India held by CSIR, New Delhi. She also cleared GATE- 2012 conducted by IIT Delhi with 180th rank in India. In August 2012, she joined Department of Chemistry, BITS Pilani, Pilani Campus under the guidance of Dr. Anil Kumar as PhD research scholar. In August, 2015 she was awarded with Senior Research Fellowship (SRF) for two years. She has published fourteen research articles in peer reviewed international journals and presented papers in four national and six international conferences with three oral presentation and seven poster presentation.



Her research interests include the development of synthetic methodologies for synthesis of bioactive heterocyclic molecule especially imidazo[1,2-*a*]pyridine and functionalization of heterocycles through transition metal-catalyzed coupling reactions involving C–H and N–H activation. She is also working on activation of C(sp³)–H bond with vanadium catalyst.

Brief Biography of the Supervisor

[A-5]

Dr. Anil Kumar is Associate Professor of Chemistry at the Birla Institute of Technology and Science, Pilani. He obtained his PhD degree from Department of Chemistry, University of Delhi, Delhi, India under the guidance of Professor SMS Chauhan in 2004. During his doctoral studies Dr. Anil Kumar worked on development of heterogeneous catalyst for organic synthesis with emphasis on green chemistry. He was postdoctoral



fellow at Department of Biomedical and Pharmaceutical Sciences, University of Rhode Island, Kingston, USA in Prof. Keykavous Parang group during May 2004 to April 2006. In his postdoctoral studies he has worked on synthesis of novel Src kinase inhibitory agents and solid phase synthesis. He joined Department of Chemistry, Birla Institute of Technology and Science, Pilani, India as Assistant Professor in 2006 and was promoted to Associate Professor in February 2013. He was appointed as Associate Dean, Work Integrated Learning Programmes (WILP) in May 2014 and Head of Department of Chemistry in September 2014. He has visited University of Rhode Island, Kingston, USA as visiting scientist and Acadia University, Wolfville, Canada as Harrison McCain visiting professor.

Dr. Kumar is recipient of Harrison McCain Foundation award from Acadia University, Canada for 2012, ISCB Young Scientist award in Chemical Sciences from Indian Society of Chemists and Biologists, Lucknow for 2013 and Dr. Aravind Kumar memorial award from Indian Council of Chemist for 2014. He has 18 year of research experience and 11 year of teaching experience. He has published over 135 research papers in international journals of repute in the area of synthetic organic chemistry, green chemistry and medicinal chemistry and contributed a book chapter and one US patent also he has written two book chapters. He has participated in several national and international symposia/conferences and delivered more than 30 invited lectures. He has guided five PhD students as supervisor and two students as co-supervisor. Currently he is supervising nine PhD students. He is editor for Canadian Chemical Transactions and member of editorial advisory board for The Open Catalysis Journal. He has completed four research projects as Principle Investigator and one as Co-PI sponsored by DST, CSIR and UGC. Currently, he has major project from DST-SERB and one industry project from Ranbaxy in collaboration with Prof. Dalip Kumar. He has also served as a reviewer for several journals. He is life member of Chemical Research Society of India, Bangalore; Indian Society of Chemists and Biologists, Lucknow and Indian Council of Chemists, Agra.

His research interest lies in transition metal catalyzed C–H activation and tandem reactions, development of reaction methodology for novel catalyst, green chemistry, design and synthesis of task specific ionic liquids and medicinal chemistry.