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Agencia de Ciencia y Tecnología  
Región de Murcia

SR. D. MEHTA VAIBHAV  
MARWADI UNIVERSITY  
RAJKOT-MORBI HIGHWAY ROAD, GAURIDAD  
360003 GUJARAT - INDIA

En el lugar y fecha de la firma electrónica

Estimado investigador:

En relación con su escrito, en el que como adjudicatario de una ayuda con cargo a la convocatoria "Ayudas a la realización de estancias de investigadores visitantes en universidades y OPIs de la Región de Murcia", integrada en el Programa Regional de Movilidad, Colaboración e Intercambio de Conocimiento "Jiménez de la Espada", en el que solicitaba poder realizar la estancia en la Universidad de Murcia desde el 3 de septiembre al 29 de noviembre, le comunico que vistas las razones expuestas, se accede a su petición.

Reciba un atento saludo

Fdo. Juan Antonio Sánchez Martínez  
Director Técnico



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C/ Manresa, 5-Entlo. 30004 Murcia  
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**Date: 20/08/2018**

**Subject: Office order to depute Dr Vaibhav Mehta as Researcher at University of Murcia from September 3, 2018 to November 29, 2018.**

**Ref: Invitation letter from Technical Director Office to Dr. Vaibhav Mehta dated 31<sup>st</sup> July 2018.**

Dr. Vaibhav Mehta is working as Professor in Faculty of Science. He has been invited as visiting researcher to work on the project “**Aids for visiting researchers to carry out stays in universities and OPIs of the Region of Murcia**” under Regional Program of Mobility, Collaboration and Knowledge Exchange, Spain during September 3, 2018 to November 29, 2018. In connection with this, he is entitled for on duty leave during the mentioned period mentioned in the subject. His travel cost, lodging and boarding will be borne by the host institute.

  
Registrar  
Marwadi University  
Rajkot

Copy to:

1. Dean and Principal of University
2. HR Department
3. Concern File



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**MEHTA, VAIBHAV PRAVINCHANDRA**

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Consulado General  
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VCESPMEHTA<<VAIBHAV<PRAVINCHANDRA<<<  
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## COMMUNICATION

## Pd-catalyzed cascade reactions involving skipped dienes: from double carbopalladation to remote C–C cleavage

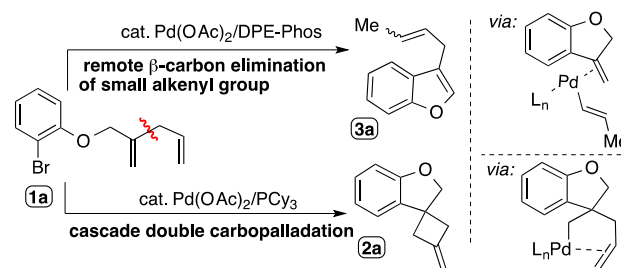
Hamid Azizollahi,<sup>a,c</sup> Vaibhav P. Mehta,<sup>b,c\*</sup> and José-Antonio García-López<sup>c\*</sup>Received 00th January 20xx,  
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We report two ligand-controlled cascade reactions relying on the intramolecular carbopalladation of skipped dienes. The use of a bulky monodentate phosphine ligand affords [4,5]-spirocycles via sequential double carbopalladation, however bidentate phosphines promote a remote  $\beta$ -C-elimination process which does not rely on the use of strained or sterically hindered substrates.

One of the driving forces that has prompted the study of new synthetic methodologies is the cleavage of ubiquitous C–C bonds.<sup>1</sup> Certainly, harnessing the routes to functionalize these linkages can boost the possibilities to use common carbon skeletons as suitable starting materials, therefore facilitating access to specific molecular substitution patterns. There are two main approaches to tackle the C–C bond functionalization by transition metals: direct C–C activation<sup>2</sup> and  $\beta$ -carbon elimination.<sup>3</sup> The cleavage of C–C bonds becomes specially challenging when they are not involved in strained systems or are located in distal positions to suitable coordinating groups. The functionalization of remote molecular moieties by means of transition metal catalysis has attracted great interest in recent years.<sup>4</sup> This field relies mainly on: a) the installation of suitable directing groups, and b) cascade reactions where an initial reactive site moves within the molecular skeleton. While these strategies have been deeply studied for the functionalization of remote C–H bonds,<sup>5</sup> they have barely been applied to remote C–C cleavage.<sup>6</sup>

Transient  $\sigma$ -alkyl Pd(II) intermediates arising from intramolecular carbopalladation of alkenes have proven to be a versatile tool for the synthesis of carbo- and heterocyclic scaffolds<sup>5b,7</sup> since they can be directly cross-coupled with a variety of reagents.<sup>8</sup> Moreover, they can perform a remote C–H activation on a pending aryl or alkyl group present in the



Scheme 1. Ligand-controlled divergent cascade from skipped dienes.

core structure, giving rise to interesting spirocycles.<sup>5,9</sup> Nevertheless, their potential to promote the C–C cleavage is underexplored.<sup>10</sup>

In 2017, Lautens and co-workers showed that unstrained  $\sigma$ -alkyl-Pd(II) species (generated via intramolecular cascade process) could effectively undergo a  $\beta$ -aryl elimination.<sup>6b</sup> In analogy to the Catellani chemistry,<sup>11</sup> the bulkiness of a sterically demanding 2,6-disubstituted arene on the  $\beta$ -carbon was the driving force of the reaction. Intrigued by these results, and in connection with our interest in the field,<sup>9,12</sup> we set to study the behaviour of 2-haloaryl-substituted ethers (**1a**, Scheme 1) bearing a skipped diene moiety toward Pd(0) catalysis.

We uncovered a ligand-controlled cascade reaction taking place on skipped dienes, which renders either [4,5]-spirocycles upon double carbopalladation of the substrate, or 3-allyl substituted benzofurans generated *via* remote C–C cleavage from an unstrained  $\sigma$ -alkyl-Pd(II) intermediate (Scheme 1).

An initial reaction of **1a** in the presence of 10 mol% of Pd(OAc)<sub>2</sub>, 20 mol% of PPh<sub>3</sub> and 1.5 equiv of Cs<sub>2</sub>CO<sub>3</sub> in toluene at 80 °C gave nearly full conversion of the starting material, affording a 1:2 mixture of the products **2a** and **3a** (Scheme 1). The [4,5]-spirocycle **2a** was the expected product arising from sequential double carbopalladation of the skipped diene followed by  $\beta$ -hydrogen elimination (path **a**, Scheme 2). Intramolecular cascade carbopalladation of alkenes constitute a powerful tool that has allowed the synthesis of [5,6]- and [6,6]-spirocyclic scaffolds.<sup>5</sup> Nevertheless they have barely been applied to the synthesis of the [4,5]-analogues,<sup>13</sup> probably due to the scarce number of published works on 4-exo-trig-

<sup>a</sup> Department of Chemistry, Faculty of Science, Ferdowsi University of Mashhad, 91775-1436, Mashhad, (Iran).

<sup>b</sup> Department of Chemistry, Marwadi University, Rajkot - Morbi Highway, Gujarat, Rajkot, 360003 (India). E-mail: mehtavp.13@gmail.com.

<sup>c</sup> Grupo de Química Organometálica, Universidad de Murcia, Campus de Espinardo, 30100, Murcia (Spain). E-mail: joangalo@um.es.

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