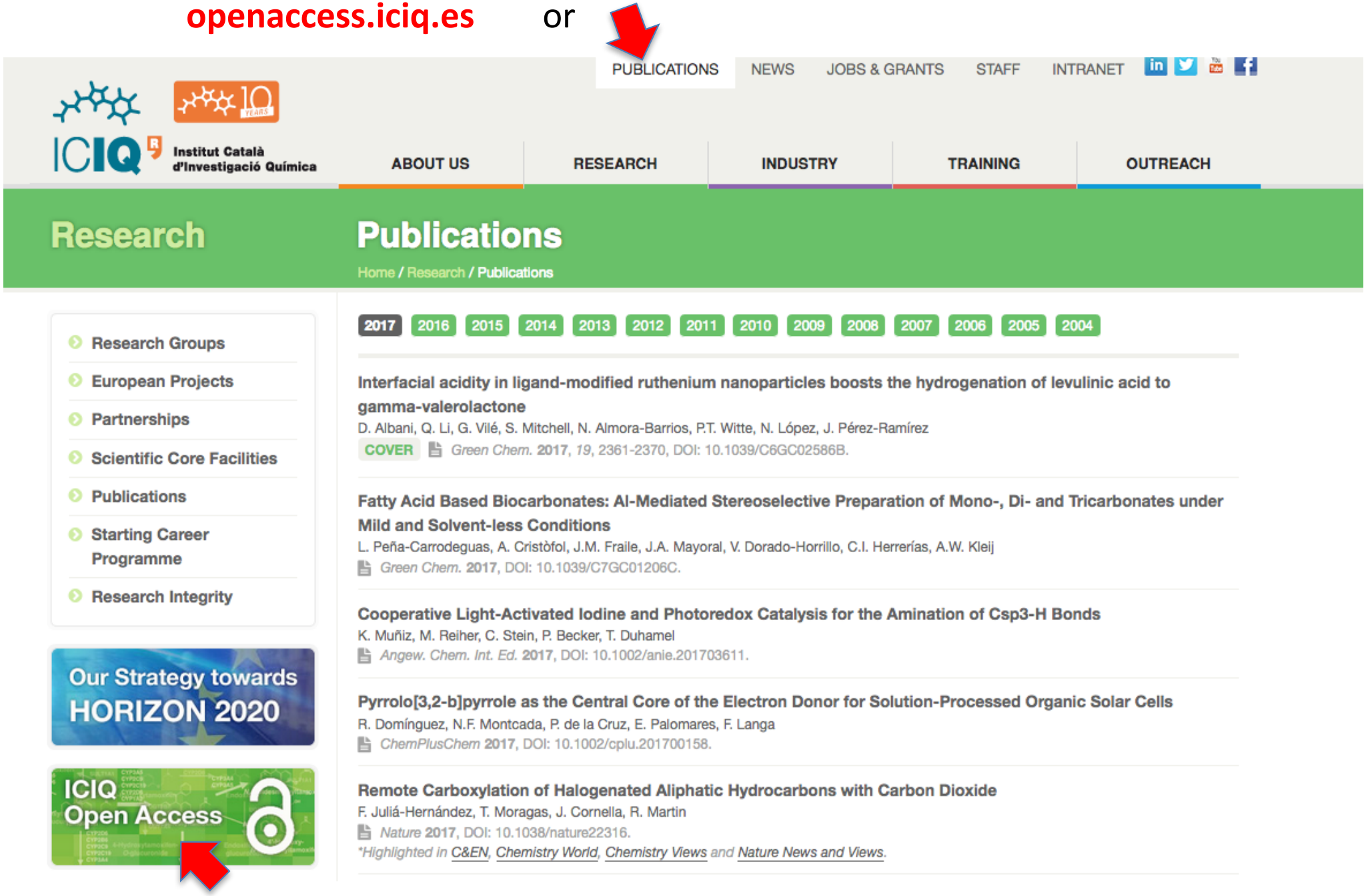
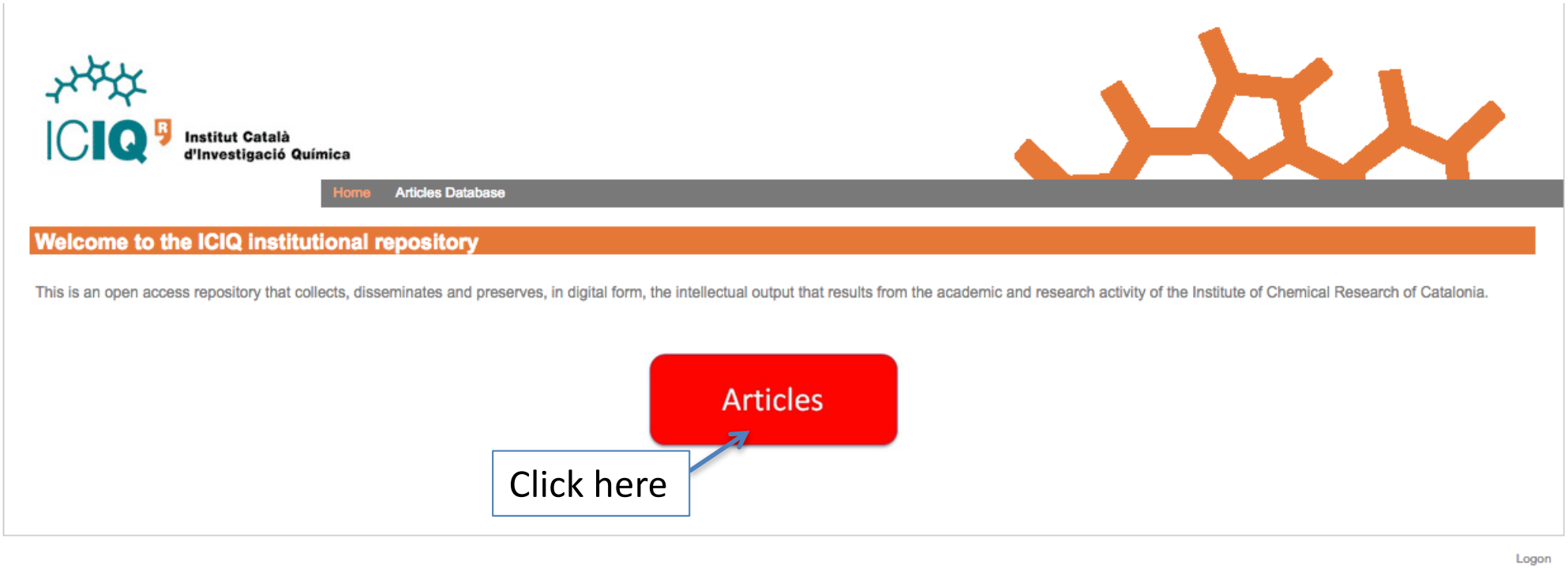
How to access to the ICIQ Institutional Repository

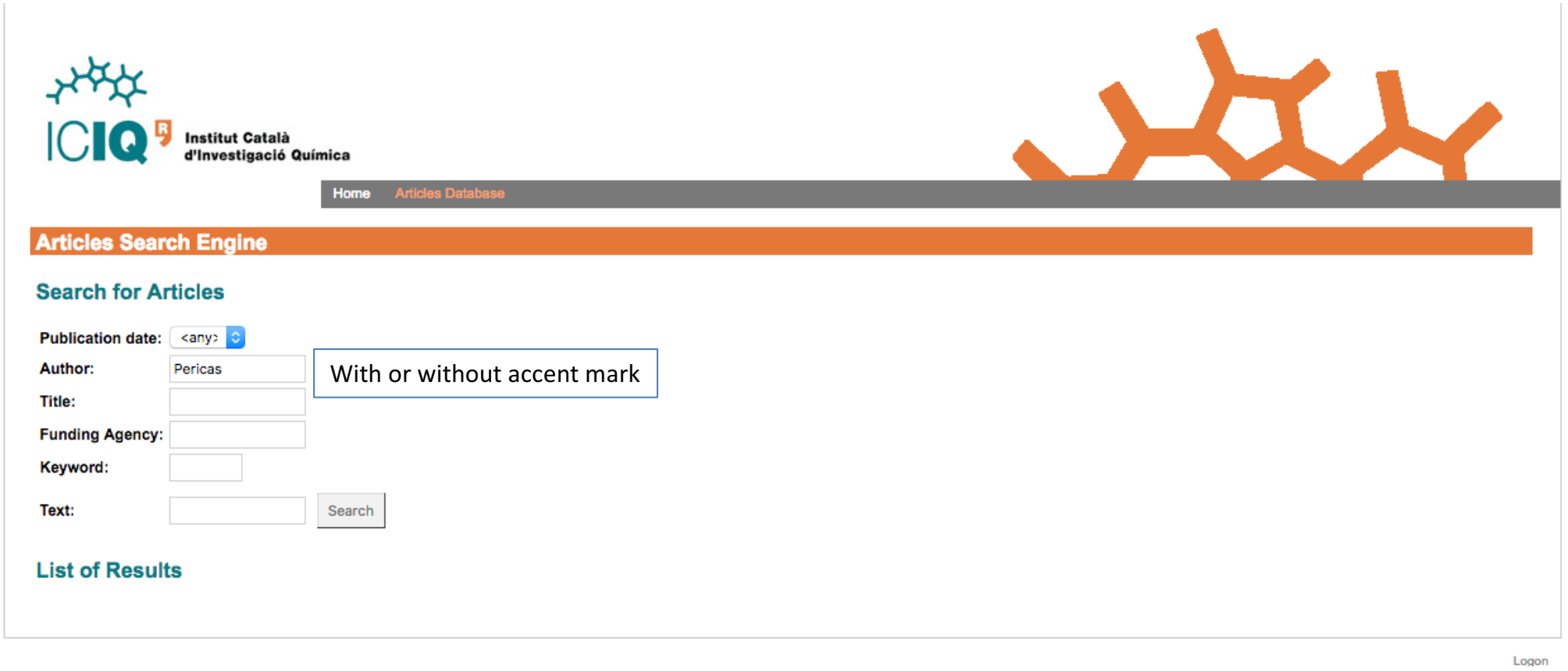
openaccess.iciq.es or



ICIQ open access database



e.g. papers by Pericàs Group: any year (2013 to date)



Logon

e.g. papers by Pericàs Group: any year (2013 to date)

Search for Articles

Publication date: <any>

Author: Pericas

Title:

Funding Agency:

Keyword:

Text: Search

Click here for article details

List of Results

Key Non-Metal Ingredients for Cu-catalyzed "Click" Reactions in Glycerol: Nanoparticles as Efficient Forwarders (2016)
Authors: Marta Rodríguez – Rodríguez Dr. Patricia Llanes, Christian Pradel Prof. Miquel A. Pericàs Prof. Montserrat Gómez
Journal: Chemistry Journal
Funding Agency: MINECO, DEC
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012–38594-C02–01, CTQ2015-69136-R, DEC 2014SGR827
Keywords:

Double-Supported Silica-Metal–Organic Framework Palladium Nanocatalyst for the Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes (2014)
Authors: Vlad Pascanu Antonio Bermejo Gómez Carles Ayats Ana Eva Platero-Prats Fabian Carson Jie Su Qingxia Yao Miquel À. Pericàs Xiaodong Zou and Belén Martín-Matute
Journal: ACS Catalysis
Funding Agency: MINECO and DEC founds
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012-38594- C02-01, 2009SGR623
Keywords: aerobic oxidation flow chemistry MIL-88B-NH2 palladium nanoparticles teabag catalysis

A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow (2017)
Authors: Santiago Cañellas Carles Ayats Andrea H. Henseler and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with FEDER founds
Action: Proyectos I+D+I – Retos 2015 Severo Ochoa Excellence Accreditation
Acronym: ROBOCAT
Grant Number: CTQ2015-69136-R, SEV-2013-0319
Keywords: continuous flow Hajos–Parrish ketone heterogeneized catalysts Robinson annulation Wieland–Miescher ketone

Metal-Free Intermolecular Azide–Alkyne Cycloaddition Promoted by Glycerol (2015)
Authors: Marta Rodríguez-Rodríguez Dr. Emmanuel Gras Prof. Miquel A. Pericàs Prof. Montserrat Gómez
Journal: Chemistry A European Journal
Funding Agency: MINECO
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012–38594-C02–01
Keywords: glycerol • azide-alkyne cycloaddition • metal-free • microwave activation • DFT modelling

Polystyrene-Supported TRIP: A Highly Recyclable Catalyst for Batch and Flow Enantioselective Allylation of Aldehydes (2016)
Authors: Lidia Clot-Almenara Carles Rodríguez-Escrich Laura Osorio-Planes and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with DEC Generalitat de Catalunya founds

PDF not available until 10/29/2017

Key Non-Metal Ingredients for Cu-catalyzed "Click" Reactions in Glycerol: Nanoparticles as Efficient Forwarders
Chem. Eur. J. 2016, 22, 18247 –18253
First published: 28 October 2016

Article details

Title:	Key Non-Metal Ingredients for Cu-catalyzed "Click" Reactions in Glycerol: Nanoparticles as Efficient Forwarders
Direct Link:	http://openaccess.iciq.es/portal/itemID=187/1090/Default.aspx
PDF Document:	The article is under embargo
Authors:	Marta Rodríguez – Rodríguez; Dr. Patricia Llanes, Christian Pradel; Prof. Miquel A. Pericàs; Prof. Montserrat Gómez
Journal:	Chemistry Journal
Publisher:	Wiley
Document Type:	Article
Additional Document Type:	Article – Peer Reviewed Version
Embargo:	10/28/2017
Funding Agency:	MINECO, DEC
Action:	Proyectos de investigación fundamental no orientada
Acronym:	SMARTCAT
Grant Number:	CTQ2012–38594-C02–01, CTQ2015-69136-R, DEC 2014SGR827
Publication Date (year):	2016
Keywords:	
Identifier (DOI):	10.1002/chem.201604048
Rights:	Copyright © 1999 - 2017 John Wiley & Sons, Inc.
Abstract:	The effect of long-alkyl-chain amines in CuI-assisted azide–alkyne cycloadditions of terminal alkynes with organic azides in glycerol and other environmentally benign solvents (water, ethanol) has been examined. The presence of these additives favors the in situ formation of Cu^{I}-based nanoparticles and results in an increase of the catalytic reactivity. In glycerol, liquid-phase transmission electron microscopy (TEM) analyses, enabled by the negligible vapor pressure of this solvent, proved that Cu^{I} nanoparticles are responsible for the observed catalytic activity. The wide variety of alkynes and azides of which this effect has been investigated (14 combinations) confirms the role played by these additives in Cu-catalyzed Huisgen cycloadditions.

In a table (provided by the ICIQ C&I Unit) you can see the embargo period policy of the journals where most of the ICIQ papers are published. Please check at the moment of publication.

Journal	**Editorial**	**Embargo period (final peer-reviewed manuscript)**
Chem. Eur. J.	Wiley	12 months

Search for Articles

Publication date: <any>
Author: Pericas
Title:
Funding Agency:
Keyword:
Text: Search

e.g. PDF available

List of Results

Key Non-Metal Ingredients for Cu-catalyzed "Click" Reactions in Glycerol: Nanoparticles as Efficient Forwarders (2016)
Authors: Marta Rodríguez – Rodríguez Dr. Patricia Llanes, Christian Pradel Prof. Miquel A. Pericàs Prof. Montserrat Gómez
Journal: Chemistry Journal
Funding Agency: MINECO, DEC
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012–38594-C02–01, CTQ2015-69136-R, DEC 2014SGR827
Keywords:

Click here for article details

Double-Supported Silica-Metal–Organic Framework Palladium Nanocatalyst for the Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes (2014)
Authors: Vlad Pascanu Antonio Bermejo Gómez Carles Ayats Ana Eva Platero-Prats Fabian Carson Jie Su Qingxia Yao Miquel À. Pericàs Xiaodong Zou and Belén Martín-Matute
Journal: ACS Catalysis
Funding Agency: MINECO and DEC founds
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012-38594- C02-01, 2009SGR623
Keywords: aerobic oxidation flow chemistry MIL-88B-NH2 palladium nanoparticles teabag catalysis

A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow (2017)
Authors: Santiago Cañellas Carles Ayats Andrea H. Henseler and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with FEDER founds
Action: Proyectos I+D+I – Retos 2015 Severo Ochoa Excellence Accreditation
Acronym: ROBOCAT
Grant Number: CTQ2015-69136-R, SEV-2013-0319
Keywords: continuous flow Hajos–Parrish ketone heterogeneized catalysts Robinson annulation Wieland–Miescher ketone

Metal-Free Intermolecular Azide–Alkyne Cycloaddition Promoted by Glycerol (2015)
Authors: Marta Rodríguez-Rodríguez Dr. Emmanuel Gras Prof. Miquel A. Pericàs Prof. Montserrat Gómez
Journal: Chemistry A European Journal
Funding Agency: MINECO
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012–38594-C02–01
Keywords: glycerol • azide-alkyne cycloaddition • metal-free • microwave activation • DFT modelling

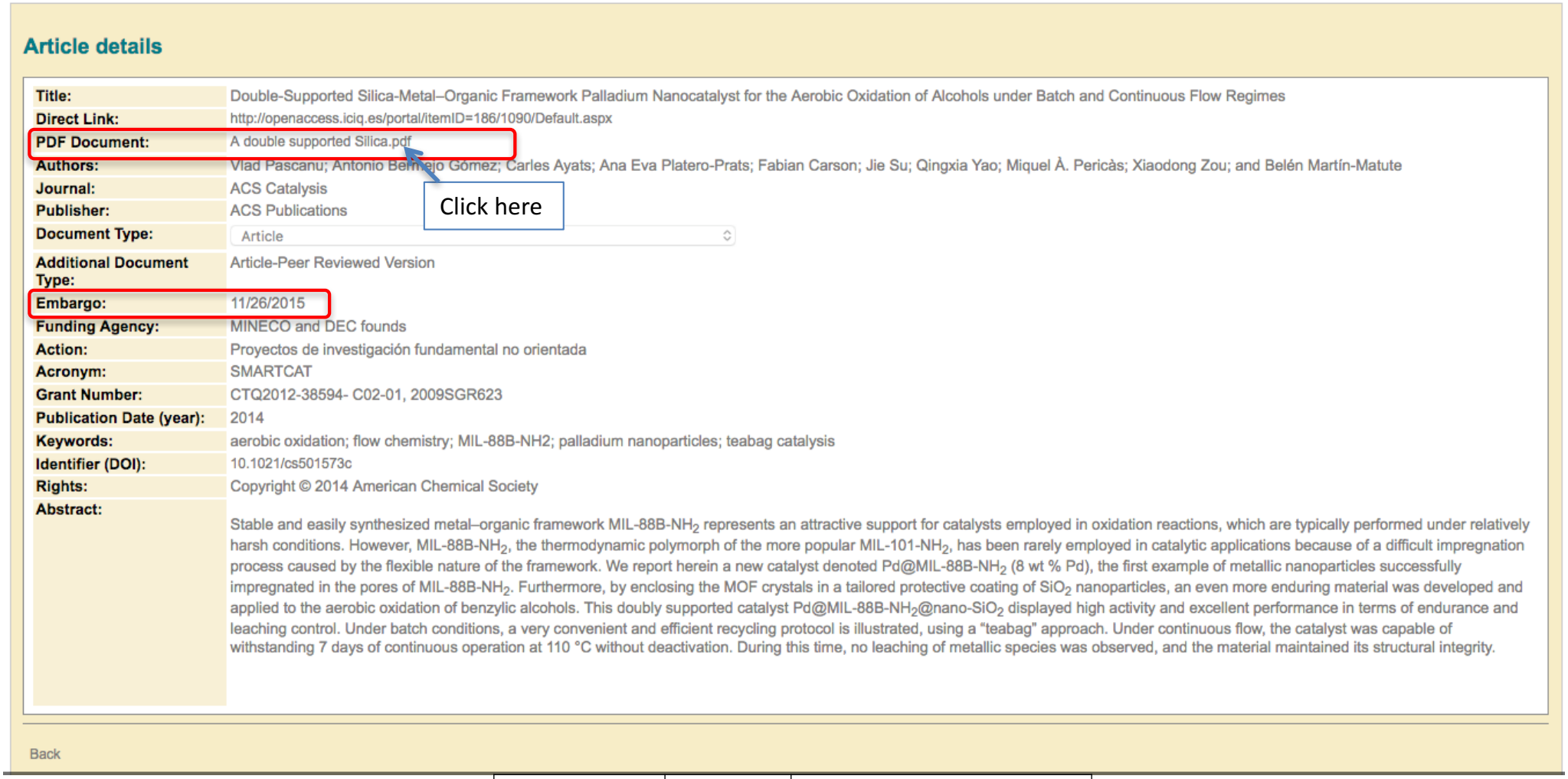
Polystyrene-Supported TRIP: A Highly Recyclable Catalyst for Batch and Flow Enantioselective Allylation of Aldehydes (2016)
Authors: Lidia Clot-Almenara Carles Rodríguez-Escrich Laura Osorio-Planes and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with DEC Generalitat de Catalunya founds

Double-Supported Silica-Metal–Organic Framework Palladium Nanocatalyst for the Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes

ACS Catal., 2015, 5 (2), pp 472–479

Publication Date (Web): November 26, 2014

e.g. PDF available



Article details

Title:	Double-Supported Silica-Metal–Organic Framework Palladium Nanocatalyst for the Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes
Direct Link:	http://openaccess.iciq.es/portal/itemID=186/1090/Default.aspx
PDF Document:	A double supported Silica.pdf
Authors:	Vlad Pascanu; Antonio Bermejo Gómez; Carles Ayats; Ana Eva Platero-Prats; Fabian Carson; Jie Su; Qingxia Yao; Miquel À. Pericàs; Xiaodong Zou; and Belén Martín-Matute
Journal:	ACS Catalysis
Publisher:	ACS Publications
Document Type:	Article
Additional Document Type:	Article-Peer Reviewed Version
Embargo:	11/26/2015
Funding Agency:	MINECO and DEC founds
Action:	Proyectos de investigación fundamental no orientada
Acronym:	SMARTCAT
Grant Number:	CTQ2012-38594- C02-01, 2009SGR623
Publication Date (year):	2014
Keywords:	aerobic oxidation; flow chemistry; MIL-88B-NH2; palladium nanoparticles; teabag catalysis
Identifier (DOI):	10.1021/cs501573c
Rights:	Copyright © 2014 American Chemical Society
Abstract:	Stable and easily synthesized metal–organic framework MIL-88B-NH_2 represents an attractive support for catalysts employed in oxidation reactions, which are typically performed under relatively harsh conditions. However, MIL-88B-NH_2, the thermodynamic polymorph of the more popular MIL-101-NH_2, has been rarely employed in catalytic applications because of a difficult impregnation process caused by the flexible nature of the framework. We report herein a new catalyst denoted Pd@MIL-88B-NH_2 (8 wt % Pd), the first example of metallic nanoparticles successfully impregnated in the pores of MIL-88B-NH_2. Furthermore, by enclosing the MOF crystals in a tailored protective coating of SiO_2 nanoparticles, an even more enduring material was developed and applied to the aerobic oxidation of benzylic alcohols. This doubly supported catalyst Pd@MIL-88B-NH_2@nano-SiO_2 displayed high activity and excellent performance in terms of endurance and leaching control. Under batch conditions, a very convenient and efficient recycling protocol is illustrated, using a "teabag" approach. Under continuous flow, the catalyst was capable of withstanding 7 days of continuous operation at 110 °C without deactivation. During this time, no leaching of metallic species was observed, and the material maintained its structural integrity.

Click here

Back

Journal	**Editorial**	**Embargo period (final peer-reviewed manuscript)**
ACS Catal.	ACS	12 months

ACS Catalysis

Research Article

pubs.acs.org/acscatalysis

Double-Supported Silica-Metal–Organic Framework Palladium Nanocatalyst for the Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes

Vlad Pascanu,[†,‡,#] Antonio Bermejo Gómez,[†,‡,#] Carles Ayats,[∥] Ana Eva Platero-Prats,[‡,§] Fabian Carson,[‡,§] Jie Su,[‡,§] Qingxia Yao,[‡,§] Miquel À. Pericàs,[∥,⊥] Xiaodong Zou,*[,‡,§] and Belén Martín-Matute*[,†,‡]

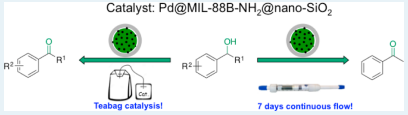
[†]Department of Organic Chemistry, [‡]Berzelii Center EXSELENT on Porous Materials, and [§]Department of Materials and Environmental Chemistry, Stockholm University, Stockholm SE-10691, Sweden

[∥]Institute of Chemical Research of Catalonia, E-43007 Tarragona, Spain

[⊥]Department of Organic Chemistry, University of Barcelona, E-08028 Barcelona, Spain

Supporting Information

ABSTRACT: Stable and easily synthesized metal–organic framework MIL-88B-NH_2 represents an attractive support for catalysts employed in oxidation reactions, which are typically performed under relatively harsh conditions. However, MIL-88B-NH_2, the thermodynamic polymorph of the more popular MIL-101-NH_2, has been rarely employed in catalytic applications because of a difficult impregnation process caused by the flexible nature of the framework. We report herein a new catalyst denoted Pd@MIL-88B-NH_2 (8 wt % Pd), the first example of metallic nanoparticles successfully impregnated in the pores of MIL-88B-NH_2. Furthermore, by enclosing the MOF crystals in a tailored protective coating of SiO_2 nanoparticles, an even more enduring material was developed and applied to the aerobic oxidation of benzylic alcohols. This doubly supported catalyst Pd@MIL-88B-NH_2@nano-SiO_2 displayed high activity and excellent performance in terms of endurance and leaching control. Under batch conditions, a very convenient and efficient recycling protocol is illustrated, using a "teabag" approach. Under continuous flow, the catalyst was capable of withstanding 7 days of continuous operation at 110 °C without deactivation. During this time, no leaching of metallic species was observed, and the material maintained its structural integrity.



KEYWORDS: *palladium nanoparticles, MIL-88B-NH_2, aerobic oxidation, teabag catalysis, flow chemistry*

INTRODUCTION

The aerobic oxidation of alcohols is an important industrial process. Sustained interest in the field is partially driven by the prospect of applying aerobic oxidation to biomass conversion, which is an abundant source of alcohols.[1] Recent advances in the area aim to use mild conditions and replace hazardous reagents with air as the oxidant.[2] Supported heterogeneous oxidation catalysts[3] have also received widespread attention because they can be recovered and recycled,[4] and palladium is often the transition metal chosen to perform the oxidation reaction.[5] For example, Pd@ARP (amphiphilic resin)[6] was used for the aerobic oxidation of secondary alcohols in water, and Pd@SBA-15 (SBA, functionalized Santa Barbara amorphous silica material)[7] was used to oxidize benzylic alcohols at temperatures as low as 80 °C. Hutchings and co-workers reported that Au–Pd bimetallic core–shell nanoparticles supported on TiO_2 could selectively oxidize benzylic and allylic alcohols with exceptional turnover frequencies (269 000 h^{-1}) under neat conditions at 100–160 °C.[8]

Metal–organic frameworks (MOFs) are porous crystalline materials assembled from metallic ions or clusters and organic multitopical linkers.[9] MOFs have gained popularity as supports in heterogeneous catalysis[10,11] because of their high surface areas and large pore volumes, which can accommodate the active catalytic sites. Another advantage is the possibility to modify MOFs postsynthetically in order to tune the properties of the framework.[12] Metallic nanoparticles can be stabilized in the pores of MOFs through both mechanical and charge-transfer interactions.[13] Through these mechanisms, highly active, small nanoparticles with a narrow size distribution can be generated, and their leaching can be avoided, facilitating the recycling of the catalyst. On the basis of these properties, combining the benefits of MOFs and flow chemistry can be regarded as a natural direction for progress in the field, and we recently reported the first example of metallic nanoparticles supported on MOFs for flow processes.[11d]

Among the first MOF-supported catalysts employed for the oxidation of alcohols, Li and co-workers developed a Pd catalyst supported on MIL-101-NH_2.[14] This system displayed good efficiency if molecular oxygen was bubbled through the reaction

Received: October 14, 2014
Revised: November 24, 2014
Published: November 26, 2014

PDF from the journal website

PDF from the ICIQ repository

The version posted must include a notice on the first page (see the recommendations provided by the ICIQ C&I Unit)

A Double–Supported Silica – Metal-Organic Framework Palladium Nanocatalyst for Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes

Vlad Pascanu,[a,c§] Antonio Bermejo Gómez,[a,c§] Carles Ayats,[d] Ana Eva Platero-Prats,[b,c] Fabian Carson,[b,c] Jie Su,[b,c] Qingxia Yao,[b,c] Miquel À. Pericàs,[d,e] Xiaodong Zou,[b,c*] Belén Martín-Matute[a,c*]

[a] Department of Organic Chemistry, Stockholm University, Stockholm, SE-10691, Sweden.

[b] Department of Materials and Environmental Chemistry, Stockholm University, Stockholm, SE-10691, Sweden.

[c] Berzelii Center EXSELENT, Stockholm University, Stockholm, SE-10691, Sweden.

[d] Institute of Chemical Research of Catalonia, E-43007, Tarragona, Spain.

[e] Department of Organic Chemistry, University of Barcelona, E-08028 Barcelona, Spain.

ABSTRACT: The stable and easily synthesized metal-organic framework MIL-88B-NH_2 represents an attractive support for catalysts employed in oxidation reactions, which are typically performed under relatively harsh conditions. However, MIL-88B-NH_2, the thermodynamic polymorph of the more popular MIL-101-NH_2, has been rarely employed in catalytic applications due to a difficult impregnation process caused by the flexible nature of the framework. We report herein a new catalyst denoted Pd@MIL-88B-NH_2 (8 wt% Pd), the first example of metallic nanoparticles successfully impregnated in the pores of MIL-88B-NH_2. Further, by enclosing the MOF crystals in a tailored protective coating of SiO_2 nanoparticles, an even more enduring material was developed and applied to the aerobic oxidation of benzylic alcohols. This double supported catalyst Pd@MIL-88B-NH_2@nanoSiO_2, displayed high activity and an excellent performance in terms of endurance and leaching control. Under batch conditions, a very convenient and efficient recycling protocol is illustrated, using a "teabag" approach. Under continuous flow, the catalyst was capable to withstand 7 days of continuous operation at 110 C, without deactivation. During this time, no leaching of metallic species was observed and the material maintained its structural integrity.

KEYWORDS: Palladium Nanoparticles, MIL-88B-NH_2, Aerobic Oxidation, Teabag catalysis, Flow Chemistry.

Introduction

The aerobic oxidation of alcohols is an important industrial process. The sustained interest in the field is partially driven by the prospect of applying aerobic oxidation to biomass conversion, which is an abundant source of alcohols.[1] Recent advances in the area aim to use mild conditions and replace hazardous reagents with air as the oxidant.[2] Supported heterogeneous oxidation catalysts[3] have also received widespread attention because they can be recovered and recycled,[4] and palladium is often the transition metal chosen to perform the oxidation reaction.[5] For example, Pd@ARP (amphiphilic resin)[6] was used for the aerobic oxidation of secondary alcohols in water and Pd@SBA-15 (SBA = functionalized Santa Barbara Amorphous silica material) was used to oxidize benzylic alcohols at temperatures as low as 80 °C. Hutchings and co-workers reported that Au-Pd bimetallic core-shell nanoparticles supported on TiO_2 could selectively oxidize benzylic and allylic alcohols with exceptional turnover frequencies (269000 h^{-1}) under neat conditions at 100–160 °C.[8]

Metal–organic frameworks (MOFs) are porous crystalline materials assembled from metallic ions or clusters and organic multi-topical linkers.[9] MOFs have gained popularity as supports in heterogeneous catalysis[10,11] because of their high surface areas and large pore volumes, which can accommodate the active catalytic sites. Another advantage is the possibility to post-synthetically modify MOFs in order to tune the properties of the framework.[12] Metallic nanoparticles can be stabilized in

A Recyclable, Immobilized Analogue of Benzotetramisole for Catalytic Enantioselective Domino Michael Addition/Cyclization Reactions in Batch and Flow (2016)

Authors: Javier Izquierdo Miquel A. Pericàs

Journal: ACS Catalysis

Funding Agency: MINECO DEC Generalitat de Catalunya

Action: Severo Ochoa Excellence Accreditation 2014−2018 Proyectos I+D+I Marie Skłodowska-Curie COFUND

Acronym:

Grant Number: SEV-2013-0319 CTQ2012-38594-C02-01 PCOFUND-GA-2011-291787 (ICIQ-IPMP) 2014 SGR827

Keywords: asymmetric catalysis, isothioureas, polystyrene-supported catalysts, domino reactions, continuous flow

e.g. Open Access paper

Article details

Title:	A Recyclable, Immobilized Analogue of Benzotetramisole for Catalytic Enantioselective Domino Michael Addition/Cyclization Reactions in Batch and Flow
Direct Link:	http://openaccess.iciq.es/portal/itemID=46/1090/Default.aspx
PDF Document:	acscatal2E5b02121.pdf
Authors:	Javier Izquierdo; Miquel A. Pericàs
Journal:	ACS Catalysis
Publisher:	ACS Publications
Document Type:	Article
Additional Document Type:	open access article

ACS Catalysis

Research Article

pubs.acs.org/acscatalysis

A Recyclable, Immobilized Analogue of Benzotetramisole for Catalytic Enantioselective Domino Michael Addition/Cyclization Reactions in Batch and Flow

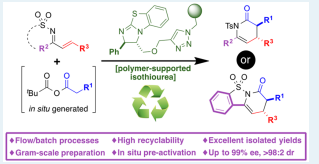
Javier Izquierdo[†] and Miquel A. Pericàs[*,†,‡]

[†]Institute of Chemical Research of Catalonia (ICIQ), The Barcelona Institute of Science and Technology, Avinguda Països Catalans 16, 43007 Tarragona, Spain

[‡]Departament de Química Orgànica, Universitat de Barcelona (UB), 08028 Barcelona, Spain

Supporting Information

ABSTRACT: A polystyrene-supported, enantiopure benzotetramisole (BTM) analogue (**5**) has been synthesized from (2*S*,3*S*)-phenylglycidol through a five-step sequence involving a copper-catalyzed alkyne azide cycloaddition (CuAAC) reaction as the final, immobilization step. The functional resin **5** (f = 0.9 mmol g^{-1}) has been successfully used as a highly active and enantioselective catalyst in the domino Michael addition/cyclization reaction of in situ activated arylacetic acids (**7**) with chalcone-type tosylimines (**6**), leading to dihydropyridinones **8** (17 examples) to afford the products with excellent yields and very high enantioselectivity (mean ee 97.4%). The deactivation of **5** by species present during the catalytic process has been studied, and pivaloyl chloride (required for the activation of the arylacetic acid) has been identified as the main source of deactivation. A simple experimental protocol taking this fact into account has allowed the multiple recycling of **5** with only a marginal decrease in catalytic activity and the implementation of a continuous flow process where the activation of phenylacetic acid (residence time 14.2 min), the asymmetric domino Michael addition/cyclization reaction (residence time 7.5 min), and aqueous workup are performed sequentially, delivering the dihydropyridinone product as a clean dichloromethane solution (0.54 mmol catalyst sample; 11 h operation; **8a** (4.44 g, >99.9% ee)). The supported catalyst **5** has also been used in a new domino Michael addition/cyclization reaction involving saccharin-derived tosylimines **9** as electrophiles, leading to 8,9-dihydro-7*H*-benzo[4,5]isothiazolo[2,3-*a*]pyridin-7-one 5,5-dioxides **10** (8 examples) in high isolated yields and diastereoselectivities and excellent enantioselectivities (mean ee 98%). A single sample of **5** (0.5 g, 0.45 mmol) has been used for the sequential preparation in batch of a library of 7 different derivatives **10** at the gram scale (ca. 10 g, accumulated TON = 51), the whole process being performed without any column chromatographic purification. The increased diastereoselectivity recorded with **5** in reactions involving sterically congested arylacetic acids (with respect to homogeneous BTM) has been rationalized through the occurrence of steric interactions between the sulfonylimine and the linker plus support catalyst fragments leading to additional destabilization of the transition state leading to the minor, cis diastereomer of products **8**/**10**.



KEYWORDS: *asymmetric catalysis, isothioureas, polystyrene-supported catalysts, domino reactions, continuous flow*

1. INTRODUCTION

Organocatalysts present inherent practical advantages, such as tolerance to moisture or oxygen, high availability (as many organocatalysts are derived from abundant natural products), and low or no toxicity. In addition, organocatalysis provides simple and effective pathways for transformations hardly achievable by other methodologies.[1] For all these reasons, organocatalysis is called upon to play an increasingly important role in key industrial sectors, including the pharmaceutical industry. When this new perspective is considered, aspects of organocatalysis such as reduced catalyst loadings, easy recovery, and the possibility of repeated recycling come to the front not only for sustainability but also for economic reasons.

The use of chiral immobilized catalysts by the synthetic community is experiencing a continued increase due to the significant advantages provided by this strategy: straightforward catalyst separation and recovery can be achieved by a simple filtration (polymer matrices) or magnetic decantation (magnetic nanoparticles) with suppression of wasteful workup stages. In addition, sufficiently active immobilized catalytic systems allow the implementation of a flow process for the continuous production of chiral enantiopure compounds.[2]

Received: September 23, 2015
Revised: December 2, 2015
Published: December 4, 2015

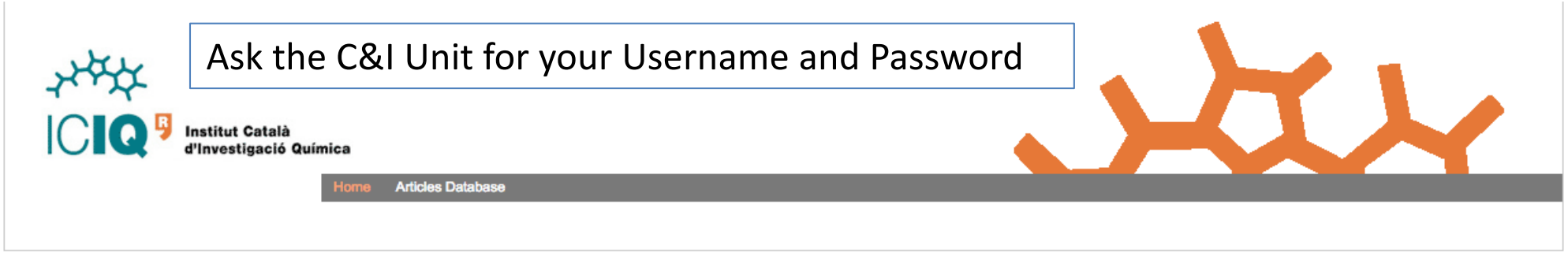
DOI: 10.1021/acscatal.5b02121
ACS Catal. 2016, 6, 348−356

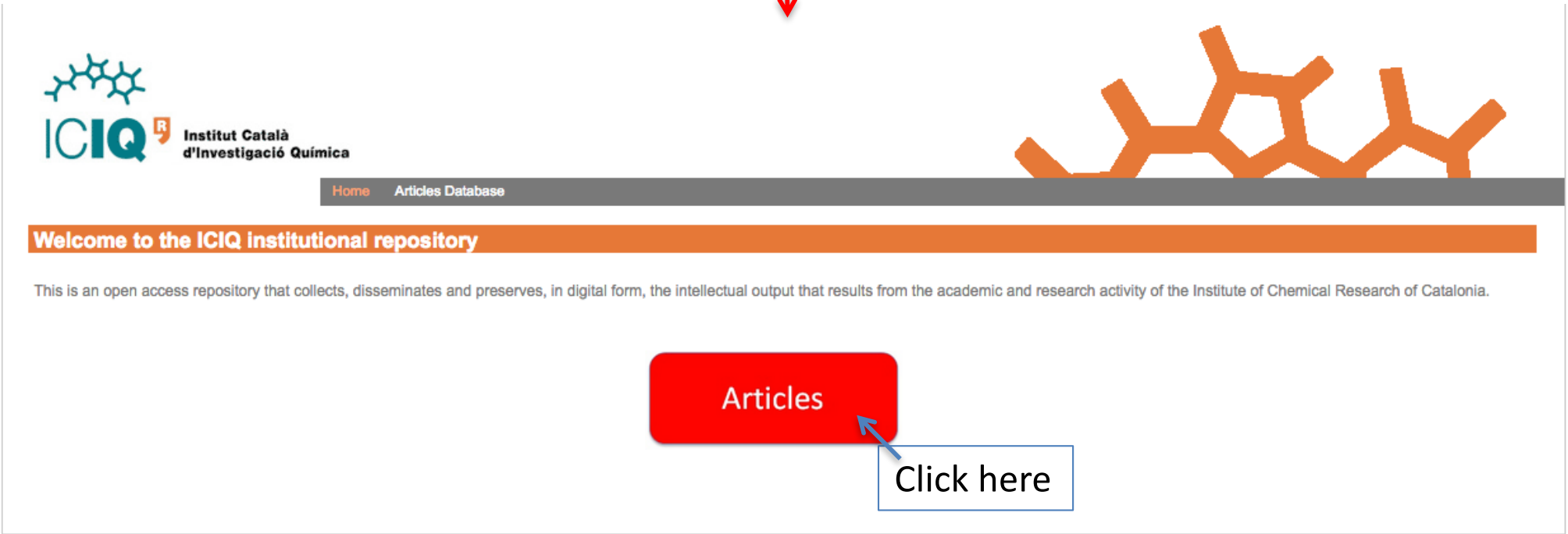
Access to the database as user

Logon

Click here

Ask the C&I Unit for your Username and Password





Search for Articles

Publication date: <any>
Author: Pericas
Title:
Funding Agency:
Keyword:
Text: Search

You can also access from the “Search” page by clicking on “Logon”

List of Results

Key Non-Metal Ingredients for Cu-catalyzed "Click" Reactions in Glycerol: Nanoparticles as Efficient Forwarders (2016)
Authors: Marta Rodríguez – Rodríguez Dr. Patricia Llanes, Christian Pradel Prof. Miquel A. Pericàs Prof. Montserrat Gómez
Journal: Chemistry Journal
Funding Agency: MINECO, DEC
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012–38594-C02–01, CTQ2015-69136-R, DEC 2014SGR827
Keywords:

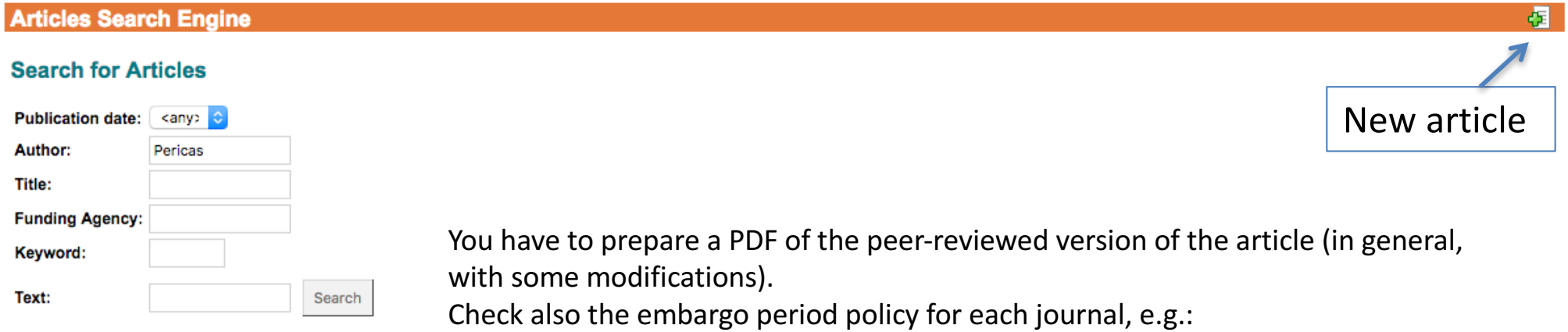
Double-Supported Silica-Metal–Organic Framework Palladium Nanocatalyst for the Aerobic Oxidation of Alcohols under Batch and Continuous Flow Regimes (2014)
Authors: Vlad Pascanu Antonio Bermejo Gómez Carles Ayats Ana Eva Platero-Prats Fabian Carson Jie Su Qingxia Yao Miquel À. Pericàs Xiaodong Zou and Belén Martín-Matute
Journal: ACS Catalysis
Funding Agency: MINECO and DEC founds
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012-38594- C02-01, 2009SGR623
Keywords: aerobic oxidation flow chemistry MIL-88B-NH2 palladium nanoparticles teabag catalysis

A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow (2017)
Authors: Santiago Cañellas Carles Ayats Andrea H. Henseler and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with FEDER founds
Action: Proyectos I+D+I – Retos 2015 Severo Ochoa Excellence Accreditation
Acronym: ROBOCAT
Grant Number: CTQ2015-69136-R, SEV-2013-0319
Keywords: continuous flow Hajos–Parrish ketone heterogeneized catalysts Robinson annulation Wieland–Miescher ketone

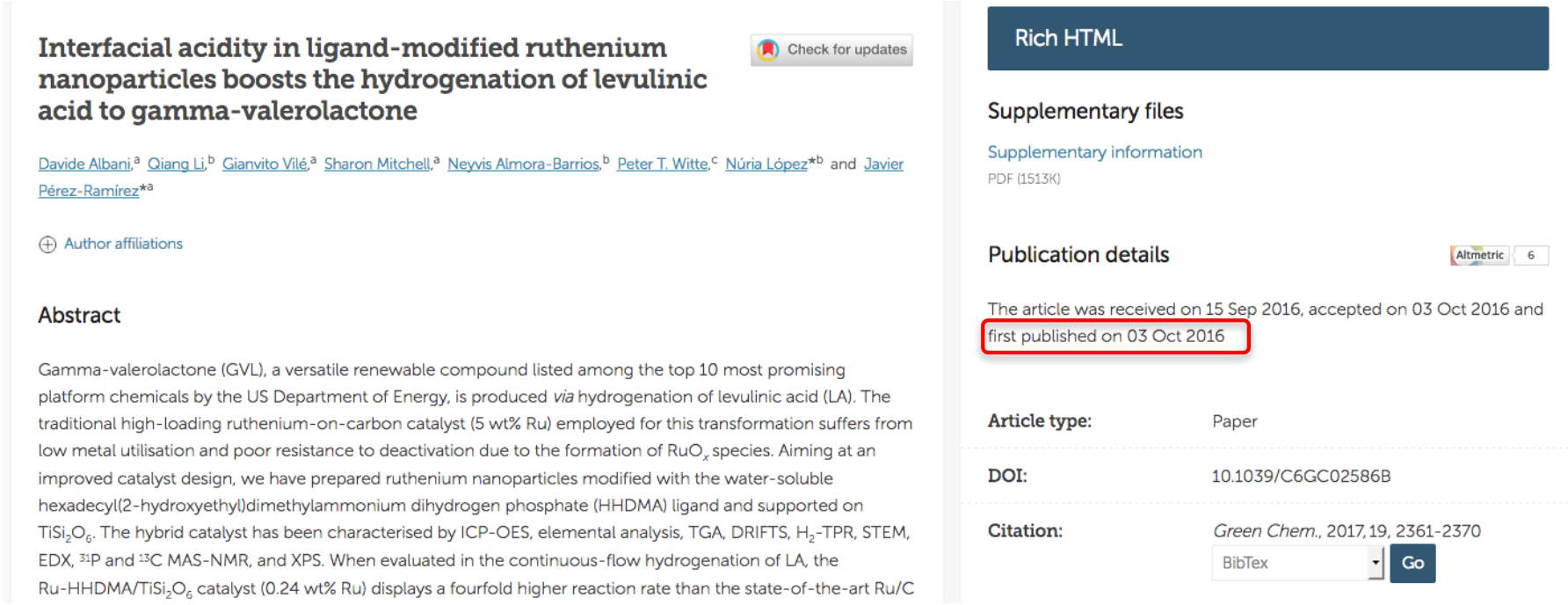
Metal-Free Intermolecular Azide–Alkyne Cycloaddition Promoted by Glycerol (2015)
Authors: Marta Rodríguez-Rodríguez Dr. Emmanuel Gras Prof. Miquel A. Pericàs Prof. Montserrat Gómez
Journal: Chemistry A European Journal
Funding Agency: MINECO
Action: Proyectos de investigación fundamental no orientada
Acronym: SMARTCAT
Grant Number: CTQ2012–38594-C02–01
Keywords: glycerol • azide-alkyne cycloaddition • metal-free • microwave activation • DFT modelling

Polystyrene-Supported TRIP: A Highly Recyclable Catalyst for Batch and Flow Enantioselective Allylation of Aldehydes (2016)
Authors: Lidia Clot-Almenara Carles Rodríguez-Escrich Laura Osorio-Planes and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with DEC Generalitat de Catalunya founds

Logon



You have to prepare a PDF of the peer-reviewed version of the article (in general, with some modifications).
Check also the embargo period policy for each journal, e.g.:



Journal	Editorial	Embargo period (final peer-reviewed manuscript)
Green Chem.	RSC	12 months

You have to prepare a PDF of the peer-reviewed version of the article (in general, with some modifications)

ChemComm

FEATURE ARTICLE

View Article Online
View Journal | View Issue

Synthesis and catalytic applications of C_3-symmetric tris(triazolyl)methanol ligands and derivatives

Pablo Etayo,[a] Carles Ayats[a] and Miquel A. Pericàs*[ab]

Cite this: *Chem. Commun.*, 2016, 52, 1997

Received 28th October 2015,
Accepted 16th December 2015

DOI: 10.1039/c5cc08961a

www.rsc.org/chemcomm

Recently introduced tris(1,2,3-triazol-4-yl)methanols and derivatives (TTM ligands) have become a valuable subclass of C_3-symmetric tripodal ligands for transition metal-mediated reactions. TTM-based ligand architectures are modularly constructed through regioselective, one-pot triple [3+2] cycloaddition of azides and alkynes. Applications of homogeneous systems of this type and of heterogenised (polystyrene- and magnetic nanoparticle-supported) TTM ligands in synthesis and catalysis are compiled in this Feature Article.

1. Introduction and general remarks

The 1*H*-1,2,3-triazole scaffold is absent from natural structures; however, this heterocyclic unit finds ubiquitous presence in myriad synthetic compounds. In the early 60's of the last century, the discovery by Huisgen of the 1,3-dipolar thermal cycloaddition of azides and alkynes[1] triggered the first synthetic approaches towards triazole heterocycles. Nonetheless, it was not until the early 2000s, when the ground-breaking reports by the groups of Meldal,[2] and Fokin and Sharpless[3] on the copper-catalysed azide–alkyne cycloaddition (CuAAC) reactions for the highly efficient, regioselective synthesis of 1,4-disubstituted 1,2,3-triazoles (Fig. 1) provided a definitive impulse to this area.[2,3] Extensive research on [3+2] cycloadditions of azides and alkynes prompted the discovery of the ruthenium-catalysed variant (RuAAC)[4] of the reaction, enabling synthetic access to 1,5-disubstituted 1,2,3-triazole regioisomers in a selective manner. Nowadays, the synthesis of 1,4-disubstituted 1,2,3-triazoles *via* CuAAC reactions[5] is considered as the paradigmatic example of click chemistry.[6] Thus, the process (Fig. 1) delivers in a predictable manner the target nitrogen-containing heterocycles (**C**) from their precursors (**A** and **B**) under mild conditions and with practically no limitations with respect to the functional groups on the reactants.

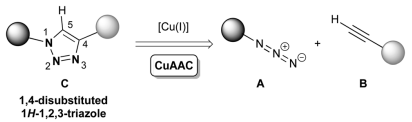


Fig. 1 Retrosynthetic approach to 1,4-disubstituted 1,2,3-triazoles *via* CuAAC.

Since its discovery, the CuAAC reaction has become a very commonly employed ligation tool. Given its reliability, specificity and biocompatibility, the CuAAC reaction has propelled the development of countless applications for triazole chemistry in diverse disciplines from pharmaceutical to materials science. Accordingly, 1,4-disubstituted 1,2,3-triazoles have emerged as valuable, yet readily available building blocks in the preparation of functional materials displaying an enormous wealth of unique properties.[7]

Regarding biomedical and biological applications, the profound impact of click reactions and triazole products in the vast field of medicinal chemistry has been extensively reviewed.[8] The triazole chemical inertness together with its inherent structural features converts this heterocycle into an excellent amide-bond surrogate (bioisosteric replacement). Consistently, triazole rings have been inserted into peptide sequences for the development of peptidomimetics that mimic the secondary structure of proteins, a topic of high relevance in chemical biology.[9] Another closely related important application of the CuAAC reaction arises from its bioorthogonality (*i.e.*, non-interacting with biological components while proceeding under physiological conditions, namely in aqueous medium at ambient temperature). This last feature has enabled the conjugation of several biomacromolecules – encompassing proteins, nucleic acids, lipids and glycans – with biophysical probes delivering biocompatible systems for both *in vitro* and *in vivo* studies.[10]

Concerning the preparation of triazole-based molecular architectures, CuAAC reactions have exerted a tremendous

[a] *Institute of Chemical Research of Catalonia (ICIQ), The Barcelona Institute of Science and Technology, Avgda. Països Catalans 16, E-43007 Tarragona, Spain. E-mail: mapericas@iciq.es; Fax: +34 977920244; Tel: +34 977920243*
[b] *Departament de Química Orgànica, Universitat de Barcelona, 08028 Barcelona, Spain*

PDF from the journal website

Journal Name

Dynamic Article Links

Cite this: DOI: 10.1039/c0xx00000x

www.rsc.org/xxxxxx

ARTICLE TYPE

Synthesis and Catalytic Applications of C_3-Symmetric Tris(triazolyl)methanol Ligands and Derivatives

Pablo Etayo,[a] Carles Ayats[a] and Miquel A. Pericàs*[a,b]

Received (in XXX, XXX) Xth XXXXXXXXX 20XX, Accepted Xth XXXXXXXXX 20XX
DOI: 10.1039/b000000x

Recently introduced tris(1,2,3-triazol-4-yl)methanols and derivatives (TTM ligands) have become a valuable subclass of C_3-symmetric tripodal ligands for transition metal-mediated reactions. TTM-based ligand architectures are modularly constructed through regioselective, one-pot triple [3+2] cycloaddition of azides and alkynes. Applications of homogeneous systems of this type and of heterogenized (polystyrene- and magnetic nanoparticle-supported) TTM ligands in synthesis and catalysis are compiled in this *Feature Article*.

1. Introduction and general remarks

The 1*H*-1,2,3-triazole scaffold is absent from natural structures; however, this heterocyclic unit finds ubiquitous presence in myriad synthetic compounds. In the early 60's of the last century, the discovery by Huisgen of the 1,3-dipolar thermal cycloaddition of azides and alkynes[1] triggered the first synthetic approaches towards triazole heterocycles. Nonetheless, it was not until the early 2000s, when the ground-breaking reports by the groups of Meldal,[2] and Fokin and Sharpless[3] on the copper-catalysed azide–alkyne cycloaddition (CuAAC) reactions for the highly efficient, regioselective synthesis of 1,4-disubstituted 1,2,3-triazoles (Fig. 1) provided a definitive impulse to this area.[2,3] Extensive research on [3+2] cycloadditions of azides and alkynes prompted the discovery of the ruthenium-catalysed variant (RuAAC)[4] of the reaction, enabling synthetic access to 1,5-disubstituted 1,2,3-triazole regioisomers in a selective manner. Nowadays, the synthesis of 1,4-disubstituted 1,2,3-triazoles *via* CuAAC reactions[5] is considered as the paradigmatic example of click chemistry.[6] Thus, the process (Fig. 1) delivers in a predictable manner the target nitrogen-containing heterocycles (**C**) from their precursors (**A** and **B**) under mild conditions and with practically no limitations with respect to the functional groups on the reactants.

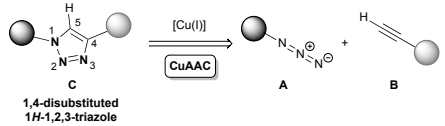


Fig. 1 Retrosynthetic approach to 1,4-disubstituted 1,2,3-triazoles *via* CuAAC.

Since its discovery, the CuAAC reaction has become a very commonly employed ligation tool. Given its reliability, specificity and biocompatibility, the CuAAC reaction has propelled the development of countless applications for triazole chemistry in diverse disciplines from pharmaceutical to materials science. Accordingly, 1,4-disubstituted 1,2,3-triazoles have emerged as valuable, yet readily available building blocks in the preparation of functional materials displaying an enormous wealth of unique properties.[7]

Regarding biomedical and biological applications, the profound impact of click reactions and triazole products in the vast field of medicinal chemistry has been extensively reviewed.[8] The triazole chemical inertness together with its inherent structural features converts this heterocycle into an excellent amide-bond surrogate (bioisosteric replacement). Consistently, triazole rings have been inserted into peptide sequences for the development of peptidomimetics that mimic the secondary structure of proteins, a topic of high relevance in chemical biology.[9] Another closely related important application of the CuAAC reaction arises from its bioorthogonality (*i.e.*, non-interacting with biological components while proceeding under physiological conditions, namely in aqueous medium at ambient temperature). This last feature has enabled the conjugation of several biomacromolecules –encompassing proteins, nucleic acids, lipids and glycans– with biophysical probes delivering biocompatible systems for both *in vitro* and *in vivo* studies.[10]

Concerning the preparation of triazole-based molecular architectures, CuAAC reactions have exerted a tremendous influence in the materials chemistry arena. Thus, the reaction has provided access to well-defined, complex polymeric materials (linear polymers, surfaces, star-shaped polymers),[11] functional polymers (stimuli-responsive hydrogels)[12] and also dendrimers.[13] Supramolecular chemistry has also benefitted from the diverse supramolecular interactions involving 1,2,3-triazoles.[14] In this context, click-derived triazoles have led to a wide range of

Peer-reviewed version of the article

This is the peer reviewed version of the following article Chem. Commun. **2016**, *52*, 1997-2010, which has been published in final form http://pubs.rsc.org/en/content/articlelanding/2016/cc/c5cc08961a#!divAbstract

Synthesis and Catalytic Applications of C_3-Symmetric Tris(triazolyl)methanol Ligands and Derivatives

Pablo Etayo,[a] Carles Ayats[a] and Miquel A. Pericàs[a,b]*

Recently introduced tris(1,2,3-triazol-4-yl)methanols and derivatives (TTM ligands) have become a valuable subclass of C_3-symmetric tripodal ligands for transition metal-mediated reactions. TTM-based ligand architectures are modularly constructed through regioselective, one-pot triple [3+2] cycloaddition of azides and alkynes. Applications of homogeneous systems of this type and of heterogenized (polystyrene- and magnetic nanoparticle-supported) TTM ligands in synthesis and catalysis are compiled in this *Feature Article*.

1. Introduction and general remarks

The 1*H*-1,2,3-triazole scaffold is absent from natural structures; however, this heterocyclic unit finds ubiquitous presence in myriad synthetic compounds. In the early 60's of the last century, the discovery by Huisgen of the 1,3-dipolar thermal cycloaddition of azides and alkynes[1] triggered the first synthetic approaches towards triazole heterocycles. Nonetheless, it was not until the early 2000s, when the ground-breaking reports by the groups of Meldal,[2] and Fokin and Sharpless[3] on the copper-catalysed azide–alkyne cycloaddition (CuAAC) reactions for the highly efficient, regioselective synthesis of 1,4-disubstituted 1,2,3-triazoles (Fig. 1) provided a definitive impulse to this area.[2,3] Extensive research on [3+2] cycloadditions of azides and alkynes prompted the discovery of the ruthenium-catalysed variant (RuAAC)[4] of the reaction, enabling synthetic access to 1,5-disubstituted 1,2,3-triazole regioisomers in a selective manner. Nowadays, the synthesis of 1,4-disubstituted 1,2,3-triazoles *via* CuAAC reactions[5] is considered as the paradigmatic example of click chemistry.[6] Thus, the process (Fig. 1) delivers in a predictable manner the target nitrogen-containing heterocycles (**C**) from their precursors (**A** and **B**) under mild conditions and with practically no limitations with respect to the functional groups on the reactants.

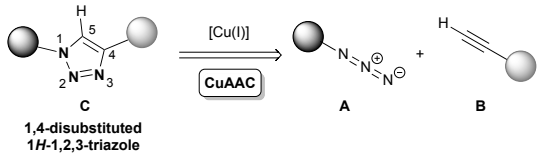


Fig. 1 Retrosynthetic approach to 1,4-disubstituted 1,2,3-triazoles *via* CuAAC.

Since its discovery, the CuAAC reaction has become a very commonly employed ligation tool. Given its reliability, specificity and biocompatibility, the CuAAC reaction has propelled the development of countless applications for triazole chemistry in diverse disciplines from pharmaceutical to materials science. Accordingly, 1,4-disubstituted 1,2,3-triazoles have emerged as valuable, yet readily available building blocks in the preparation of functional materials displaying an enormous wealth of unique properties.[7]
Regarding biomedical and biological applications, the profound impact of click reactions and triazole products in the vast field of medicinal chemistry has been extensively reviewed.[8] The triazole chemical inertness together with its inherent structural features converts this heterocycle into an excellent amide-bond surrogate (bioisosteric replacement). Consistently, triazole rings have been inserted into peptide sequences for the development of peptidomimetics that mimic the secondary structure of proteins, a topic of high relevance in chemical biology.[9] Another closely related important application of the CuAAC reaction arises from its bioorthogonality

Recommendations provided by the ICIQ C&I Unit:

Wiley journals:
"This is the peer reviewed version of the following article: [FULL CITE], which has been published in final form at [Link to final article using the DOI]. This article may be used for non-commercial purposes in accordance with Wiley Terms and Conditions for Self-Archiving."

ACS journals:
“This document is the Accepted Manuscript version of a Published Work that appeared in final form in [JournalTitle], copyright © American Chemical Society after peer review and technical editing by the publisher. To access the final edited and published work see [insert ACS Articles on Request author- directed link to Published Work, see http://pubs.acs.org/page/policy/articlesonrequest/index.html].”

Example for a RSC paper (Pericàs Group)

This is the peer reviewed version of the following article Chem. Commun. **2016**, *52*, 1997-2010, which has been published in final form
http://pubs.rsc.org/en/content/articlelanding/2016/cc/c5cc08961a#!divAbstract

Articles Search Engine

Search for Articles

Publication date: <any>

Author: Pericas

Title:

Funding Agency:

Keyword:

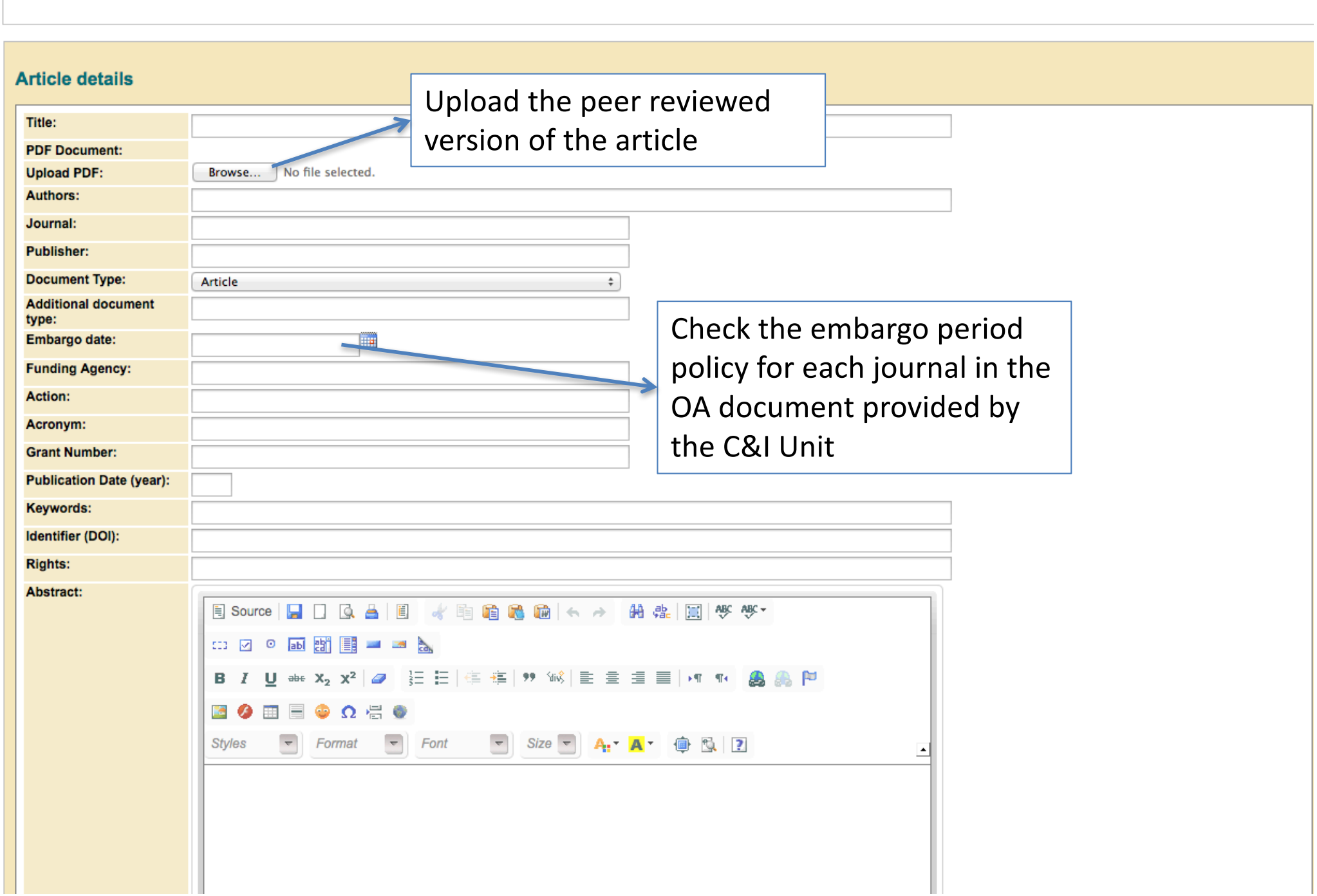
Text: Search

New article

Article details

Upload the peer reviewed version of the article

Check the embargo period policy for each journal in the OA document provided by the C&I Unit



A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow

Santiago Cañellas, Carles Ayats, Andrea H. Henseler and Miquel A. Pericàs

ACS Catal., 2017, 7 (2), pp 1383–1391
DOI: 10.1021/acscatal.6b03286
Publication Date (Web): January 10, 2017

Article details

Title:	A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow
PDF Document:	A Highly Active Polymer.pdf
Upload PDF:	Browse... No file selected.
Authors:	Santiago Cañellas; Carles Ayats; Andrea H. Henseler; and Miquel A. Pericàs
Journal:	ACS Catalysis
Publisher:	ACS Publications
Document Type:	Article
Additional document type:	Article-Peer Reviewed Version
Embargo date:	01/10/2018
Funding Agency:	MINECO with FEDER founds
Action:	Proyectos I+D+I – Retos 2015; Severo Ochoa Excellence Accreditation
Acronym:	ROBOCAT
Grant Number:	CTQ2015-69136-R, SEV-2013-0319
Publication Date (year):	2017
Keywords:	continuous flow; Hajos–Parrish ketone; heterogeneized catalysts; Robinson annulation; Wieland–Miescher ketone
Identifier (DOI):	10.1021/acscatal.6b03286
Rights:	Copyright © 2017 American Chemical Society

"Authors" separated by a semicolon

"Actions" separated by a semicolon

"Grant numbers" *should be* separated by a semicolon

"Keywords" separated by a semicolon

Abstract:

The preparation through Robinson annulation of enantiopure building blocks with both academic and industrial relevance, such as the Wieland–Miescher and Hajos–Parrish ketones, has suffered from important drawbacks, such as the need for high catalyst loading or extremely long reaction times. Here we report a heterogenized organocatalyst based on Luo's diamine for fast and broad-scope enantioselective Robinson annulation reactions. The polystyrene-supported diamine **19a** enables the high-yield, highly enantioselective preparation of a wide range of chiral bicyclic enones under mild conditions, with reaction times as short as 60 min (batch) or residence times of 10 min (flow). In contrast with its homogeneous counterpart **19b**, the catalytic resin **19a** experiences a notable increase in catalytic activity with temperature in 2-MeTHF (a 10-fold decrease in reaction times without erosion in enantioselectivity is observed from room temperature to 55 °C). The scope of the transformation in batch mode has been illustrated with 14 examples, including examples only reported in poorly enantioenriched (**22n**) or in racemic form (**22k**). Enantiopure **22k** has been used as the starting material for a straightforward formal synthesis of the antibiotic and antifeedant sesquiterpene (−)-isovelleral (**24**). The heterogenized catalyst **19a** admits extended recycling (10 cycles) and has been used to develop the first asymmetric Robinson annulations in continuous flow. The potential of the flow process is illustrated by the large-scale preparation of the Wieland–Miescher ketone (65 mmol in 24 h of operation, TON of 117) and by a sequential flow experiment leading to a library of eight enantioenriched diketone compounds.

Apply Changes | Cancel | Delete

Title:

Funding Agency:

Keyword:

Text: Search

List of Results

A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow (2017)
Authors: Santiago Cañellas Carles Ayats Andrea H. Henseler and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with FEDER founds
Action: Proyectos I+D+I – Retos 2015 Severo Ochoa Excellence Accreditation
Acronym: ROBOCAT
Grant Number: CTQ2015-69136-R SEV-2013-0319
Keywords: continuous flow Hajos–Parrish ketone heterogeneized catalysts Robinson annulation Wieland–Miescher ketone

Funding Agency: MINECO DEC Generalitat de Catalunya
Action: Severo Ochoa Excellence Accreditation 2014–2018 and Proyectos I+D+i
Acronym:
Grant Number: SEV-2013-0319 CTQ2012-38594-C02-01 2014 SGR827
Keywords: acetaldehyde · cross-aldol · recycling ·site-isolation · wolf and lamb

Logoff

Edit profile Logoff

Title:

Funding Agency:

Keyword:

Text: Search

List of Results

A Highly Active Polymer-Supported Catalyst for Asymmetric Robinson Annulations in Continuous Flow (2017)
Authors: Santiago Cañellas Carles Ayats Andrea H. Henseler and Miquel A. Pericàs
Journal: ACS Catalysis
Funding Agency: MINECO with FEDER founds
Action: Proyectos I+D+I – Retos 2015 Severo Ochoa Excellence Accreditation
Acronym: ROBOCAT
Grant Number: CTQ2015-69136-R SEV-2013-0319
Keywords: continuous flow Hajos–Parrish ketone heterogeneized catalysts Robinson annulation Wieland–Miescher ketone