Nanocatalysts from Ionic Liquid Precursors for CO_2 Valorisation to Hydrocarbons

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Introduction

The conversion of CO_2 into lower olefins (C_3-C_5) is a highly desirable process as a sustainable production route. Thereby, the use of hydrogen from renewable energies and the conversion of CO_2 into lower olefins *via* Fischer-Tropsch synthesis (FTS) offers an attractive route for efficient utilisation of biogas as a renewable feedstock to replace petroleum for the synthesis of key building-block chemicals. Lower olefins, i.e., ethylene, propylene and butene (C_2-C_4) are key building blocks in the current chemical industry. Iron-based catalysts are of interest due to their ability to catalyse both FTS and Reverse Water Gas Shift (RGS). These are also of interest as they are able to produce high olefin hydrocarbons [1]. The main reason for the iron catalyst effectiveness in such process is its formation of iron carbides (χ-Fe_5C_2) formed after reaction gas treatment. It has also been reported that the iron catalysts require alkali metal promotion in order to obtain desired activity and selectivity [2, 3]. The further upgrading to gasoline range hydrocarbons can be done by having zeolites in close proximity to the iron catalysts. It has been proposed that the zeolites crack lower chain olefins, and able to facilitate chain growth [4].However, for such catalysts, controlling the size and the particle distribution remains a major challenge. Thus, in order to obtain monodispersed catalysts, a novel approach is developed, utilising ionic liquids which can dissolve precursors while itself containing dense and tuneable network of hydrogen bonds. Such synthetic methods have been demonstrated by Wang et al. [5]. Nanoparticles produced through this method have been shown to produce higher surface areas. We report here on a novel methodology for the controlled synthesis of a Na–Fe_3O_4/HZSM-5 multifunctional catalyst for the direct hydrogenation of CO_2 to gasoline. The catalytic testing under industrially relevant conditions resulted in improved selectivity to C_5–C_{11} as well as low CH_4 and CO_2 selectivity. Furthermore, the product composition can be tuned by the zeolite properties (i.e. Si/Al ratio, H form, alkaline exchange) and by the choice of ionic liquid in the synthetic method. This study provides a new pathway for the synthesis of nanocatalysts for the production of liquid fuels by utilising CO_2 and H_2.

Materials and Methods

The ionic liquid-assisted synthesis involved heating the reaction medium consisting of a range of imidazolium based ionic liquids with different cations, oleic acid and oleylamine as the capping agents, and different iron precursors (iron pentacarbonyl, iron acetate, iron acetate, etc.) under reflux. In each instance, a black precipitate is formed. The produced magnetite nanoparticles are separated from the reaction medium through application of a neodymium magnet. The compounds were then characterised using PXRD (Fig. 1 A), SEM / EDX and TPR (Fig. 1 B).

The Na–Fe_3O_4/Zeolite catalysts were typically prepared by both powder mixing and granule mixing. It is has been observed that the iron catalyst and the zeolite proximity to each other have an effect on the selectivity [4]. For powder mixing, the Na–Fe_3O_4 catalysts with H-ZSM-5 in a ball mill at a mass ratio of the two components of 1:1. For granule mixing, the Na–Fe_3O_4 and zeolites were pressed and sized in 20-40 mesh, followed by pouring into a vessel and shaken to give the mixing.A typical CO_2 hydrogenation reactions were performed at 320 °C, 20 bar, and a 3 to 1 ratio of H_2/CO_2 in a stainless steel fixed-bed reactor with an inner diameter of 15 mm. 1 g of composite catalyst with Na–Fe_3O_4/Zeolite 1/1 (mass ratio) was used. Prior to reaction, the catalyst was reduced in pure H_2 flow at 350 °C and atmospheric pressure for 8 h. All products from the reaction is analysed with an online gas chromatograph (GC) with FID and TCD detection.

Results and Discussion

The synthesis of a nanocrystalline magnetite using ionic liquids is shown to facilitate controlled precipitation due to their dual functionality as solvent and templating agent. Additional capping agent further induces shape selectivity resulting in smaller particle size. This level of control over the morphology of the produced iron nanoparticles allows for the selectivity of the hydrocarbon distribution to be directly tailored to light olefin production. Characterization of the prepared catalysts by PXRD (Fig. 1) shows phase purity for the magnetite obtained from the conventional precipitation method, and shows no alteration after granulation with the zeolite, showing the presence of high purity in Fe_3O_4, small particle size and good dispersion with the zeolite component. The compounds obtained by the ionic liquid methods result in much broader XRD patterns, pointing at less crystallinity and smaller particle size. This has also been confirmed by SEM and TEM. Hydrogen temperature-programmed reduction (H_2-TPR) was used to determine the interaction between Fe species and the support. As shown in Figure 2 for the HZSM 80, all the catalysts present two peaks with increasing reduction temperature, which are assigned to the conversions Fe_3O_4—FeO and FeO—Fe, respectively. It is observed that Fe_3O_4/HZSM80 becomes reduced at a lower temperature compared to the catalyst with low (l) and high (H) Na content. This indicates the interaction between iron oxides and the support is weaker if Na is completely removed.

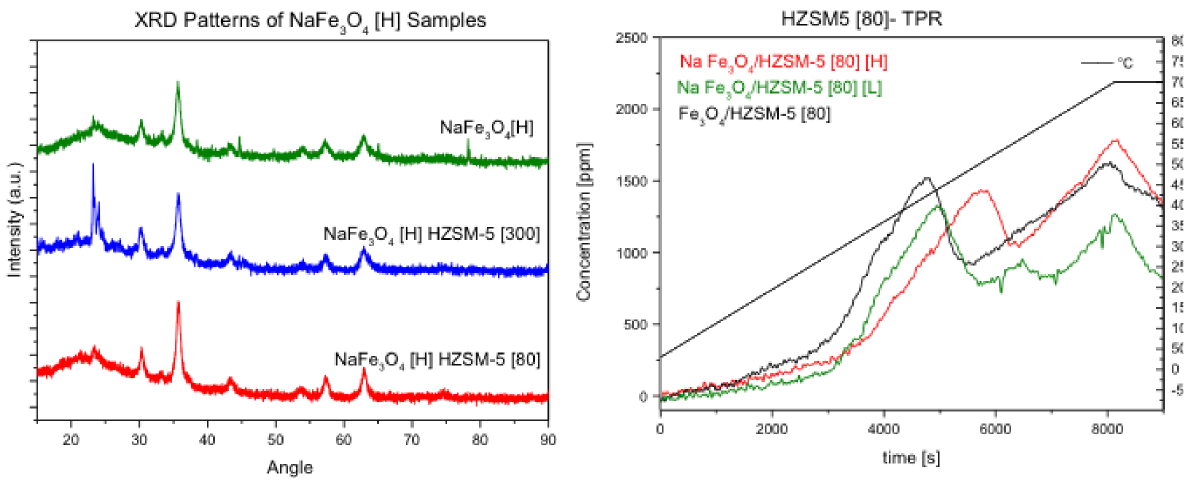

Figure 1. Panel A: XRD patterns of Na–Fe_3O_4 sample with high Na content (H) and Na–Fe_3O_4 mixed with HZSM with SiO_2/Al_2O_3 = 80 and 300.   Panel B: TPR profile of 4000 ppm H_2 in Ar from RT to 700ºC, 5ºC/min

References

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Table of Contents

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Plenary Speakers

Tim Bugg

'Bacterial Enzymes for Lignin Degradation'

Tim Bugg is Professor of Biological Chemistry at the University of Warwick. His academic career started at the University of Southampton in 1991, where his group studied enzymes involved in the bacterial degradation of aromatic compounds and enzymes involved in bacterial peptidoglycan assembly. Since moving to Warwick in 1999, his group has more recently studied enzymes involved in bacterial degradation of lignin, and the application of biocatalysis to convert lignin into renewable aromatic chemicals. He is the author of the undergraduate textbook “Introduction to Enzyme and Coenzye Chemistry”.

Alessandra Quadrelli

'Surface Organometallic Chemistry on MOFs, POPs and Inorganic Oxides for CO2 and N2 Reduction: En route to Renewable Energies Storage'

Alessandra is director of research of the French National Centre for Scientific Research, CNRS, at the nanochemistry platform of the C2P2 labs. She also chairs the CPE Lyon Engineering School Sustainable Development Chair and is Associate Editor of the RSC journal “Green chemistry”.

Her research focuses on developing molecular understanding of the interaction between organometallic precursors and solid surfaces of SiO2, MOFs and 2D wafers (among other solids). She applies this understanding to the synthesis of heterogeneous catalysts and thin films aimed at renewable energy utilization. She considers her Top-3 professional achievements: A new mechanism for N2 cleavage (SCIENCE, 2007), the creation of the “CO2 forum” conferences (http://co2forum.cpe.fr) and the synthesis of a MoS2 monolayer by Atomic Layer Deposition, ALD (NANOSCALE, 2017).

Enrico Tronconi

'The NH3-SCR Redox Cycle over Cu-CHA: Insights from Transient Response Methods'

Enrico Tronconi is a Professor of Chemical Engineering at the Department of Energy of Politecnico di Milano, Italy. His research interests concern the applications of Catalytic Reaction Engineering to environmental protection and energy conversion. Enrico has investigated DeNOx aftertreatment technologies during the last twenty years. He is also active in the study of novel structured catalysts and reactors for process intensification.

Programme

Monday, 7 September 2020

*Click on the Session number on the left to get more details of each session and talks or on the Poster session to view the posters in that session

09:00		**Welcome**			
	Session 1	Session 1A Emission Control Zoom link 333514	Session 1B Energy Zoom link 756439	Session 1C Water Zoom link 582802	
		Chair: Dr Nancy Artioli	Chair: Dr Chunfei Wu	Chair: Dr Xiaolei Fan	
		M. Keenan 16	KEYNOTE	X. OU 140	
		T. Franken 64	J. Huang 245	M. da Silva 255	
		M. Roger 78	B. Ipek Torun 12	**KEYNOTE**	
		U. Iacobone 130	P. Summa 31	A. Halder 45	
10:25		Break			
10:45	Session 2	Session 2A Emission Control Zoom link 265868	Session 2B Energy Zoom link 877654	Session 2C Water Zoom link 303676	Session 2D Emission Control Zoom link 965611
		Chair: Dr Matthew Keenan	Chair: Dr Xiaoxia Ou	Chair: Prof. Fernando Pereira	Chair: Dr Weidong Zhang
		X. Yong 69	P. Da Costa 36	**KEYNOTE**	A. Lanza 198
		C. Dujardin 158	X. Chen 41	L. Lefferts 256	R. Khawaja 118
		D. Zengel 219	G. Yang 42	E. Palomares 209	K. Swirk 218
		G. Rochard 47		P. Pietrzyk 199	A. Gervasini 102
12:10		Lunch; Visit exhibition space for Hiden Analytical (Zoom link) and Magritek (Zoom link)			
13:10		Poster session			
13:50		Opening Remarks Zoom link Passcode: 274811			
14:05		**Plenary Speaker - Dr. Tim Bugg - Bacterial Enzymes for Lignin Degradation** Chair Justin Hargreaves Zoom link Passcode: 274811			
14:50		Break			
15:05	Session 3	Session 3A Emission Control Zoom link 532431	Session 3B Energy Zoom link 005012	Session 3C Biomass Zoom link 660918	Session 3D Circular Economy Zoom link 255130
		Chair: Prof Alexandre Goguet	Chair: Dr Bahar Ipek Torun	Chair: Dr Roberto Rinaldi	Chair: Dr Chris Parlett
		KEYNOTE	Z. Baysal 15	V. Piazza 197	V. Parvulescu 22
		L. Pettersson	X. Jia 19	A. Ismaila 234	R. Arrigo 201
		M. Agote Aran 63	A. Kokka 24	M. Fuhrer 231	J. Pui Ho Li 194
16:10		Break			
16:25	Session 4	Session 4A Emission Control Zoom link 864890		Session 4C Biomass Zoom link 697953	Session 4D Circular Economy Zoom link 855401
		Chair: Dr Miren Agote Aran		Chair: Dr Haresh Manyar	Chair: Dr Chris Parlett
		M. Machida 39		**KEYNOTE**	I. Kowalec 149
		M. Nebreda 51		R. Rinaldi	Y. Wang 123
		S. Nandi 60		K. Agrawal 211	
17.30		**End**			

ICEC
International Conference on Environmental Catalysis 2020

Tuesday, 8 September 2020

Click on the Session number on the left to get more details of each session and talks or on the Poster session to view the posters in that session

Time	Session				
09:00		**Welcome**			
	Session 1	Session 1A Emission Control Zoom link 293885	Session 1B Emission Control Zoom link 833147	Session 1C Energy Zoom link 578811	
		Chair: Prof Pascal Granger	Chair: Dr Tanja Franken	Chair: Dr Jun Huang	
		E. Palomares 207	P. Lott 190	**KEYNOTE**	
		D. Ferri 222	N. Nasello 121	Xin Tu 239	
		O. Kröcher 248	O. Bjørkedal 183	J. Spivey 81	
		F. Buttignol 33	M. Ogura 40	B. Xie 133	
10:25		Break			
10:45	Session 2	Session 2A Emission Control Zoom link 469729	Session 2B Biomass Zoom link 782956	Session 2C Energy Zoom link 311539	
		Chair: Prof Dr Oliver Kröcher	Chair: Prof Justin Hargreaves	Chair: Prof Patrick Da Costa	
		KEYNOTE	S. Le 135	S. Sun 50	
		A. Goguet 57	H. Manyar 196	C. Sun 52	
		D. Kim 132	D. Dochain 179	**KEYNOTE**	
		A. Suarez 225	W. Ouyang 204	B. Tan	
12:10		Lunch; Visit exhibition space for Hiden Analytical (Zoom link) and Magritek (Zoom link)			
13:10		Poster session			
13:50		**Plenary Speaker - Dr Alessandra Quadrelli - Surface Organometallic Chemistry on MOFs, POPs and inorganic oxides for CO2 and N2 reduction: en route to renewable energies storage** Chair: Arthur Garforth Zoom link Passcode: 430913			
14:35		Break			
14:55	Session 3	Session 3A Emission Control Zoom link 549312	Session 3B Emission Control Zoom link 325933	Session 3C Energy Zoom link 296154	Session 3D Water Zoom link 465121
		Chair: Dr Davide Ferri	Chair: Dr Thomas Sheppard	Chair: Zeynep Baysal	Chair: Dr Xiaoxia Ou
		S. Fahed 151	E. Eßer 53	S. Gu	M. Hantusch 224
		T. Pellegrinelli 101	G. Isapour 91	J. Esquius 186	L. Pirault-Roy 83
15:20		Break			
15:50	Session 4	Session 4A Emission Control Zoom link 332765	Session 4B Emission Control Zoom link 441309	Session 4C Special Session Zoom link 809731	Session 4D Water Zoom link 187022
		Chair: Prof Christophe Dujardin	Chair: Dr Eleni Kyriakidou	Chair: Prof Michael Stockenhuber	Chair: Dr Xiaoxia Ou
		M. Cortés-Reyes 147	R. Nuguid 172	**KEYNOTE**	F. Boyom-Tatchemo 44
		A. Garcia 119	X. Ye 67	C. Sykes	T. Chankhanittha 17
		M. Sourek 156	T. Sheppard 238	**KEYNOTE**	
		R. Villamaina 104		M. Wong	
17:15		**End**			

ICEC
International Conference on Environmental Catalysis 2020

Wednesday, 9 September 2020

Click on the Session number on the left to get more details of each session and talks or on the Poster session to view the posters in that session

09:00	**Welcome**			
Session 1	Session 1A Emission Control Zoom link 942206	Session 1B Energy Zoom link 023283	Session 1C General Zoom link 670769	Session 1D Circular Economy Zoom link 803504
	Chair: Prof Do Heui Kim	Chair: Dr Haresh Manyar	Chair: Prof Chris Hardacre	Chair: Prof Vasile Parvulescu
	M. Strømsheim 212	**KEYNOTE**		K. Chatzipangiotou 87
	C. Singer 54	C. Wang 252	C. Hudy 210	A. Bin Jumah 205
	L. Negahdar 105	A. Mostafa 159	J. Thompson 203	
10:05	Break			
10:25 Session 2	Session 2A Emission Control	Session 2B Energy	Session 2C General	Session 2D Circular Economy
	Zoom link 700862	Zoom link 200941	Zoom link 394442	Zoom link 904086
	Chair: Dr Jillian Thompson	Chair: Professor Xin Tu	Chair: Prof Chris Hardacre	Chair: Dr Aleksander Tedstone
	P. Helie 154	S. Xu 208	C. Parlett 61	**KEYNOTE**
	J. Schnee 191	P. Xu 253	L. Harvey 180	A. Kicherer
	M. Panchal 153		S. Chansai 213	C. Poupin 66
11:30	Lunch; Visit exhibition space for Hiden Analytical (Zoom link) and Magritek (Zoom link)			
12:30	Poster session			
13:10	**Plenary Speaker - Enrico Tronconi - The NH3-SCR redox cycle over Cu-CHA: Insights from transient response methods** Chair Nancy Artioli Zoom link Passcode: 564253			
13:55	Break			
14:15 Session 3	Session 3A Emission Control Zoom link 615202	Session 3B Energy Zoom link 829514	Session 3C Water Zoom link 034121	Session 3D Emission Control Zoom link 307471
	Chair: Dr Todd Toops	Chair: Dr Jillian Thompson	Chair: Dr Xin Gao	Chair: Dr Roberta Villamaina
	KEYNOTE	J. Chen 46	K. Zhao 14	W. Zhang
	M. Votsmeier	H. Yamashita 65	S. Nanan 4	M. Drewery 168
	N. Artioli 216		**KEYNOTE**	I. Chepurna 85
	J. Lee 73		F. Pereira 189	L. Chmielarz 127
15:40	Break			
16:00 Session 4	Session 4A Emission Control Zoom link 086881		Session 4C Water Zoom link 558279	Session 4D Emission Control Zoom link 600639
	Chair: Dr Eleni Kyriakidou		Chair: Dr Xiaolei Fan	Chair: Dr Leila Negahdar
	V. Torregrosa Rivero 162		**KEYNOTE**	M. Blažek 59
	T. Toops 187		M. Symes 13	I Meza-Trujillo 152
16.45	**Closing remarks** Zoom link 432462			

Posters

Monday 7 September Click on the title to view the poster and the Zoom link to join the session

Yixi Wang https://zoom.us/j/93332493133
Enhancement of NOx adsorption performance during adsorption-regeneration cycles over Pt/Ba/Al2O3 catalyst

Paraskevi Panagiotopoulou https://zoom.us/j/97399415584
Effect of operating conditions on the catalytic performance of supported Rh catalysts for the reaction of LPG steam reforming

Sebastián Gámez https://zoom.us/j/92504964904
Carbon Black-Polydopamine-Ruthenium composite as an efficient and recyclable boomerang catalyst for the oxidative cleavage of oleic acid

Olívia Salomé Soares https://zoom.us/j/91947639213
Tuning the surface properties of carbon supporting materials to achieve efficient Ni based catalysts for CO2 methanation

Alexandre Goguet https://zoom.us/j/93011738289
Structure Selectivity of Supported Pd nanoparticles for Catalytic NH3 Oxidation resolved using combined Operando Spectroscopy

Edidiong Asuquo https://zoom.us/j/94845971866
Evaluation of hydrothermal carbonisation of biomass wastes for production of adsorbents for Cd(II) removal from aqueous solutions

Madan Behera https://zoom.us/j/95673586731
NO reduction using Pt-zeolite catalysts in O2 and CO2 rich gas atmosphere

Alain Li https://zoom.us/j/99638543980
Haemoglobin as a bio-derived precursor for FeNx single-site catalysts.

Luke Roebuck https://zoom.us/j/93843924326
Rare-Earth Doped Ceria-Zirconia Nanodispersions: Oxygen Storage Materials for Gasoline Particulate Filters

Theodora Ramantani https://zoom.us/j/95216791843
Hydrogen production by steam reforming of propane over supported noble metal catalysts

Aidan Doyle https://zoom.us/j/94588987681
Simultaneous abatement of NO and N2O with CH4 over modified Al2O3 supported Pt,Pd,Rh

Daniela Pietrogiacomi https://zoom.us/j/93672747929
Oxidative dry reforming of methane for syngas production: a promising activity of Ni/ZrO2 catalysts

Lioudmila Nossova https://zoom.us/j/95479118807
Co- and Zr-doped barium cerate perovskite catalyst for simultaneous NOx storage and soot oxidation

Lan Lan https://zoom.us/j/91450421871
Effect of ball-milling and plasma treatment on microcrystalline cellulose on the H2 production via cellulose photoreforming

Anna Szelwicka https://zoom.us/j/92209247222
CNTs-based biocatalysts dedicated for sustainable chemical processes

Return to programme

Tuesday 8 September Click on the title to view the poster and the Zoom link to join the session

Maria Ruggeri https://zoom.us/j/94645535603
Mechanistic insight in NO trapping on Pd/Chabazite systems for the low-temperature NOx removal from Diesel exhausts

Aleksandra Borcuch https://zoom.us/j/91333867881
Fe-exchanged MWW derivatives as catalysts of NH3-SCR process

Aneta Święs https://zoom.us/j/99716227131
Catalytic performance of modified ferrierites as effective catalysts for catalytic reduction of NO with ammonia

Nicolaas van Strien https://zoom.us/j/91282813973
Unique pathway to platform chemicals - 2,5-furandicarboxylic acid and muconic acid from sugar acids

Guangtao Chai https://zoom.us/j/97965913153
Effect of zironium on catalytic combustion of vinyl chloride over Co3O4-based catalysts

Tamara Kharlamova https://zoom.us/j/93846412069
Ceria-supported Pt–Ag bimetallic catalysts for CO oxidation and hydrogenation of nitrophenol

Małgorzata Sieradzka https://zoom.us/j/92460388107
Investigation of solid catalysts based on alkaline earth metals and transition metals within gasification process of biomass wastes.

Bomin Fu https://zoom.us/j/91536046859
Effects of ions and humic acid on the removal of pemetrexed in water by activated carbons

Yulia Belik https://zoom.us/j/92297661611
Effect of preparation method on photocatalytic activity of Bi-based composites in RhB and phenol photodegradation

Marina Cortés-Reyes https://zoom.us/j/95851147134
Transient Response Method to delve into NOx removal process using a hybrid NSR-SCR system

Maria Smyrnioti https://zoom.us/j/96208398254
CO oxidation in the presence of water and methanol vapor over FexCo1-x mixed oxides

Sofia Santos https://zoom.us/j/97480302366
Catalytic reduction of inorganic species formed during ozonation of organic contaminants

Return to programme

Wednesday 9 September Click on the title to view the poster and the Zoom link to join the session

Juan Carlos Martínez-Munuera https://zoom.us/j/97134478636
Unraveling the nature of active sites onto copper/ceria-zirconia catalysts for low temperature CO oxidation

Cyril Thomas https://zoom.us/j/98488294172
Exceeding the 2 wt% Ag Loading Frontier on Al2O3 for C3H6-SCR: Insights into the identification of the Al2O3 sites of importance

Alessandra Beretta https://zoom.us/j/94602322471
Enhanced kinetics of NH3-SCR in the presence of HCl in the flue gas over V-based catalysts: investigation by activity and characterization experiments (link to poster not available)

Adrian Mizera https://zoom.us/j/97269257932
Catalysts based on Ni/Co/Cu system doped with strontium titanate for dry reforming of methane

Maria Cristina Campa https://zoom.us/j/93599370865
Fe-MOR catalysts for the abatement of N2O and NOx: effect of the preparation method

Lucy Costley-Wood https://zoom.us/j/94324016471
Long Term Aging of Ceria Zirconia for Exhaust Catalyst Applications (link to poster not available)

Jose Castanheiro https://zoom.us/j/95339827460
Acetalization of glycerol with hexanal in the presence of SBA-15 with sulfonic acid groups.

Francesca Varsano https://zoom.us/j/96847770536
Innovative materials to drive chemical reactions by induction heating

Shangchao Xiong https://zoom.us/j/99329030071
The poisoning mechanism of gaseous HCl on low-temperature SCR catalysts: MnOx−CeO2 as an example

Wenhao Yang https://zoom.us/j/96190986164
Controllable Redox-induced In-situ Growth of MnO2 over Mn2O3 for Toluene Oxidation: Active Heterostructure Interfaces

Luke Forster https://zoom.us/j/98353944418
Tailoring textural properties for tuning diffusion behaviour of alumina catalytic materials: A rational guideline exploiting bench-top Pulsed-Field Gradient (PFG) Nuclear Magnetic Resonance (NMR)

Carmine D'Agostino https://zoom.us/j/92613773212
Supported organocatalysis as a greener alternative in the production of fine chemicals

Melissa Greta Galloni https://zoom.us/j/99461520533
Activity and stability of copper and iron exchanged hydroxyapatite catalysts in NH3-SCR

Zhipeng Qie https://zoom.us/j/98788729667
Trace potassium assisted catalytic activation: A scalable production method regulating pore configuration of activated coke for synergistic removal of VOCs, SO 2 and NO

Suk Bong Hong https://zoom.us/j/99774541256
Effect of preparation method on the NH3-SCR activity of Cu-LTA catalysts

Return to programme