Novel synthesis approaches for CO2 Hydrogenation catalysts using Ionic Liquids

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Introduction

The conversion of carbon dioxide into lower olefins (C2-C4) represents a highly desirable process for establishing a sustainable production pathway[1]. These lower olefins, including ethylene, propylene, and butenes, play pivotal roles in the chemical industry and the production of Liquefied Petroleum Gas (LPG). The reaction unfolds through two consecutive primary processes: Reverse Water Gas Shift (RWGS), generating CO, followed by the subsequent transformation of CO into hydrocarbons through the Fischer–Tropsch reaction[2]. Recent research has underscored the cost-effectiveness and satisfactory performance of Fe-based catalysts in both reaction steps, with an exploration of bimetallic catalysts, particularly combinations of Ru and Fe, aimed at enhancing olefin selectivity[3]. Precise synthesis of multi-nanoparticle (MNP) becomes a critical factor for performance control in this context.

The study introduces an innovative approach to synthesize iron-ruthenium bimetallic catalysts, utilizing ionic liquids as solvents[4]. This method ensures the precise and uniform distribution of active metal phases. Advanced characterizations and extensive tests reveal that this technique outperforms traditional colloid-based methods, resulting in superior selectivity for the desired hydrocarbons.

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Materials and Methods

In the traditional colloidal synthesis (COL) process[3], Ru nanoparticles (Ru-NPs) were generated using a modified Schlenk technique, employing $Ru_3(CO)_{12}$ and oleylamine at 543 K. Likewise, Fe nanoparticles were created through the use of oleylamine and $Fe(acac)_3$ at 573 K. Bimetallic Ru-Fe nanoparticles were formulated in hexane as a comparative benchmark. In the ionic liquid (IL) method, [BmIm][BF_4] was employed with either $Fe(acac)_3$ or $Ru_3(CO)_{12}$ at temperatures of 523 K for a duration of 18 hours. Subsequently, nanoparticles were recovered, and for bimetallic Fe-Ru NPs, three distinct molar ratios (1:1, 3:1, and 9:1) were investigated using $Fe(acac)_3$ and $Ru_3(CO)_{12}$ in [BmIm][PF_6] at 523 K for 18 hours. The nanocatalysts were supported on γ-Al_2O_3 with varying metal loadings (1 or 4 wt.%). Characterization involved XRF, XRD, SEM, and H_2 chemisorption. Kinetic experiments were conducted at 593 K and 6 or 20 bar pressures, adjusting the gas-hourly space velocity (GHSV) to maintain CO_2 conversion below 5%. FT-IR gas analysis was employed to assess yield and selectivity.

Results and Discussion

In **Figure 1**, TPR profiles of catalysts 1wt% Fe-Ru 3:1/Al_2O_3 (COL) and 1 wt% Fe-Ru 3:1/Al_2O_3 (IL) are presented. Both exhibit a low-temperature reduction peak, around 380°C, associated to the reduction of Fe_2O_3 to Fe_3O_4, and an high-temperature peak, starting at 620°C, indicates further reduction to FeO and Fe^0. The reduction peak for Fe_2O_3 in RuO_2-Fe_2O_3 catalysts shifts to lower temperatures compared to literature values for pure Fe_2O_3[5], that suggests easier reduction of RuO_2 species, showcasing the acceleration of Fe species reduction and hydrogen spillover from Ru to Fe_2O_3. The cooperative effect between RuO_2 and Fe_2O_3 enhances reduction properties, correlating with improved catalytic performance.

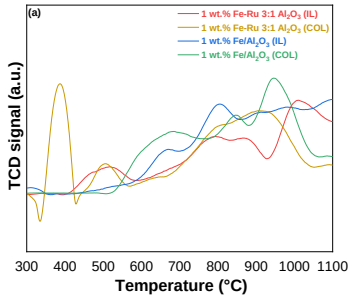

Figure 1. H_2-TPR profiles of catalysts 1 wt% Fe-Ru 3:1/Al_2O_3 (COL and (IL)).

Notably, at low temperatures (300°C < T < 470 °C), the colloidal catalyst shows a pronounced peak for the reduction of Ru^{4+} to Ru^0, while this peak diminishes in the IL catalyst, attributed to a more isolated ruthenium metal phase in the COL catalyst and a closer proximity synergistic effect in the IL catalyst. The reduction at 500 °C, linked to RuO_2 reduction associated with Fe, is more prominent in the ionic liquid catalyst. **Figure 2a** COL and **Figure 2b** IL reveal distinct selectivity profiles using catalysts via COL and IL methods. Generally, the COL catalyst exhibits a preference for CO, whereas the IL catalyst tends to shift towards CH_4 and C2-C5 hydrocarbons, whether considering monometallic or bimetallic catalysts. This selectivity shift intensifies with a higher metal loading (4 wt%).

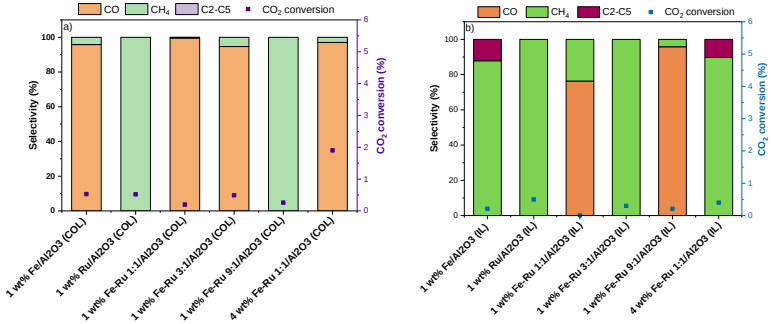

Figure 2. Hydrocarbon selectivity and CO_2 conversion at 20 bar (3:1 H_2:CO_2, 523 K, 45 mL/min)

Significance

In summary, this research highlights the superior performance of bimetallic Fe-Ru species synthesized via the IL method over conventional colloidal synthesis. This advancement holds promise for sustainable energy solutions, including CO_2 conversion into net-zero e-fuels and its use as a carbon feedstock for renewable resources.

References

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UKCC 2024

10th UK Catalysis Conference, 3-5 January 2024
Loughborough, UK

Wednesday, 3rd January			
11:00	Registration desk opens at Burleigh Court Hotel		
12:30	Lunch at Holywell Park		
13.50	Welcome – Conference commences at Holywell Park		
	Chair – Prof. Chris Hardacre		
14.00	PI 01 – Prof. Richard Catlow *(Turing Lecture Theatre)*		
14.45	Coffee		
	Session A (Turing Lecture Theatre) CatalysisHub session	*Session B (Brunel/Murdoch Lecture Theatre)*	*Session C (Stephenson Lecture Theatre)*
Chair/IT	Beale/Centeno	Kondrat/Mazumdar	Garforth/Inrirai
15.15	K1 (Weller)	O4	O11
15.35		O5	O12
15.55	O1	O6	O13
16.15	O2	O7	K2 (Matam)
16.35	O3	O8	
16.55	Coffee		
Chair/IT	Artioli/Maddaloni	Lennon/Wilding	Wang/ Nieva De La Hidalga
17.25	K3 (Fey)	O9	O14
17.45		O10	O15
18.10	Careers Question Time – *(Turing Lecture Theatre)*		
20.00	Dinner		

Thursday, 4th January			
	Chair – Prof. Graham Hutchings		
9.00	PI 02 – Prof. Silvia Bordiga *(Turing Lecture Theatre)*		
	Session A *(Turing Lecture Theatre)*	*Session B* *(Brunel/Murdoch Lecture Theatre)*	*Session C* *(Stephenson Lecture Theatre)*
Chair/IT	Mitchell/Olsen RSC INTEREST GROUP APPLIED CATALYSIS session	Simons/Asad	Petkov/Collins
10.00	K4 (Zhang)	O18	O28
10.20		O19	O29
10.40	O16	O20	O30
11.00	Coffee		
Chair/IT	Paterson/Ross	Matam/Mazumdar	Delarmelina/Maddaloni
11.30	K5 (Gibson)	O21	O31
11.50		O22	O32
12.10	O17	O23	O33
12.30	Lunch		
	Chair – Prof. Richard Catlow		
14.00	PI 03 – RSC Award Lecture – *(Turing Lecture Theatre)*		
14.45	Coffee		
	(Turing Lecture Theatre)	*(Brunel/Murdoch Lecture Theatre)*	*(Stephenson Lecture Theatre)*
Chair/IT	Mulholland/Centeno RSC INTEREST GROUP SURFACE REACTIVITY & CATALYSIS session	Garforth/Mohammad	Weller/Inrirai
15.15	K6 (Artioli)	O24	O34
15.35		O25	O35
15.55	K7 (Hermans)	O26	O36
16.15		O27	O37
16.35	Coffee		
17.00 to 19.00	**Poster session**		
20.00	Conference Dinner		

Friday, 5th January			
	Session A (Turing Lecture Theatre)	*Session B (Brunel/Murdoch Lecture Theatre)*	*Session C (Stephenson Lecture Theatre)*
Chair/IT	Freakley/Centeno	Zhang/Mohammad	Lin/Olsen
9.00	K8 (Nastase)	O40	O46
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9.40	O38	O42	O48
10.00	Coffee		
Chair/IT	Dingwall/Ross	Fey/Asad	D'Agostino/Collins
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10.50		O44	O50
11.10	O39	O45	O51
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11.35	PI 04 – Prof. Walter Leitner *(Turing Lecture Theatre)*		
12.20	Closing remarks		

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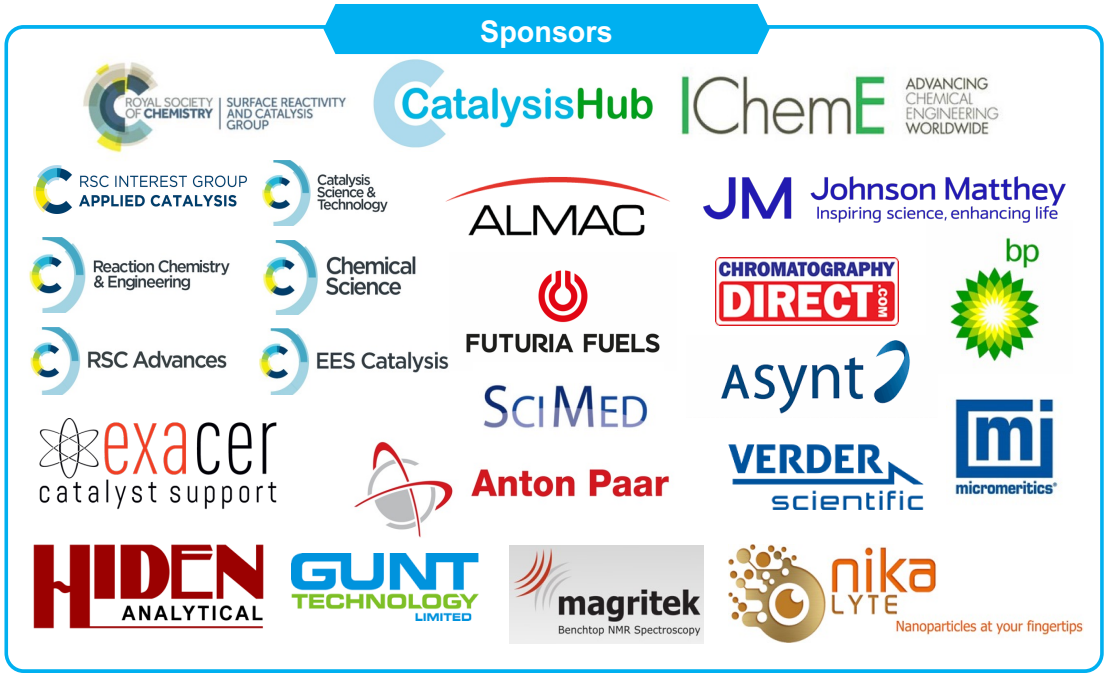

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Dr. Paul Collier, Johnson Matthey

PLENARY AND KEYNOTE SPEAKERS

UKCC 2024 will feature a number of plenary and keynote presentations from leaders across all areas of catalysis.

PLENARY SPEAKERS

Prof. Walter Leitner

Max Planck Institute for Chemical Energy Conversion, Germany

New Carbon Sources for the Energetic and Chemical Value Chain: Challenges and Opportunities for Catalysis - TEN YEARS AFTER!

Prof. Sir Richard Catlow

Cardiff Catalysis Institute, UK

Modelling of Catalytic Structures and Mechanisms: Achievements and Challenges

Prof. Silvia Bordiga

University of Turin, Italy

MOFs and MOFs derivatives used as catalysts

KEYNOTE SPEAKERS

Dr. Nancy Artioli, University of Brescia, Italy and Queen's University Belfast, UK

Dr. Natalie Fey, University of Bristol, UK

Dr. Emma Gibson, University of Glasgow, UK

Prof. Ive Hermans, University of Wisconsin-Madison, USA

Dr. Santhosh Matam, Cardiff University, UK

Dr. Stefan Nastase, King Abdullah University of Science and Technology, Saudi Arabia

Dr. James Paterson, BP, UK

Dr. Xiaodong Wang, Lancaster University, UK

Prof. Andrew Weller, University of York, UK

Dr. Xiaolei Zhang, University of Strathclyde, UK

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Pl 04	New Carbon Sources for the Energetic and Chemical Value Chain: Challenges and Opportunities for Catalysis - TEN YEARS AFTER!	Walter Leitner
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