

中国科学院院长奖申报表

申报类别 _____

姓名 张倩 _____

单位名称 中国科学院山西煤炭化学研究所 _____

学科专业 化学 _____

学科专业代码 070300 _____

攻读学位 博士研究生 _____

导师姓名及职称 樊卫斌 研究员 _____

中国科学院人才与人事局制

2026 年 04 月 08 日填

一、基本信息							
姓名	张倩	性别	女	民族	汉族	籍贯	河北省 衡水市 深州市
出生年月	1994.10.25	政治面貌	中国共产党预备党员		入学时间	2023.03.01	
办公电话		手机号码	13831896923		电子邮件	zhangqian203@mails.ucas.ac.cn	
何时何单位何专业获何种学位		2016年9月-2020年6月 在河北科技大学获得学士学位; 2020年9月-至今 在中国科学院大学获得博士学位					
个人简历	起止年月	学习和工作单位			获何种学位		
	2016年9月-2020年6月	河北科技大学			学士学位		
	2020年9月-至今	中国科学院大学			博士学位		
学位课成绩（请附成绩单）							
学年学期		课程名称		学位课	学时	学分	成绩
2024—2025 学年(秋) 第一学期		中国马克思主义与当代		是	36	2.0	87
2023—2024 学年春季学期		Seminar		是	42	3.0	83
2023—2024 学年春季学期		催化剂制备技术		是	42	3.0	93
2020—2021 学年(秋) 第一学期		知识产权		否	20	1.0	89
2020—2021 学年(秋) 第一学期		中国特色社会主义理论与实践研究		是	36	2.0	81
2020—2021 学年(秋) 第一学期		硕士学位英语		是	64	3.0	78
2020—2021 学年(秋) 第一学期		纳米科学与技术		是	30	2.0	89
2020—2021 学年(秋) 第一学期		固体表面物理化学		是	50	3.0	90
2020—2021 学年(秋) 第一学期		化学反应工程		否	50	3.0	85
2020—2021 学年(秋) 第一学期		工业催化原理		是	50	3.0	76

2020—2021 学年 (秋) 第一学期	煤化学与煤转化利用	否	50	3.0	86
2020—2021 学年 (秋) 第一学期	能源战略管理	否	36	1.0	89
2020—2021 学年 (秋) 第一学期	学术道德与学术写作规范	是	20	1.0	87
2020—2021 学年 (秋) 第一学期	工程伦理	否	20	1.0	通过
2020—2021 学年 (春) 第二学期	自然辩证法概论	是	36	1.0	87
2020—2021 学年 (春) 第二学期	博士学位英语	是	64	2.0	66
2020—2021 学年 (春) 第二学期	专业英语	否	32	2.0	80
2020—2021 学年 (春) 第二学期	环境催化和功能材料	是	50	3.0	91
2020—2021 学年 (春) 第二学期	催化反应工程	是	50	3.0	95
2020—2021 学年 (春) 第二学期	催化过程的研究范式	是	40	2.0	94
2020—2021 学年 (春) 第二学期	科技信息检索与利用实用技巧 (化学化工领域)	否	30	1.0	88
2020—2021 学年 (春) 第二学期	人文系列讲座	否	20	1.0	通过
2020—2021 学年 (夏) 第三学期	拉曼光谱在科学研究中的应用	否	20	1.0	通过
2020—2021 学年 (夏) 第三学期	生态与环境毒理展望	否	20	1.0	82
2020—2021 学年 (夏) 第三学期	科学前沿 (1)	否	20	1.0	通过
总学分	49.0	学位课 学分	33.0		
论文题目或成果 名称	CO ₂ 加氢经“低碳醇”中间体制低碳烯烃催化剂设计与构筑				
论文 (或成果) 性 质	基础研究				
二、申报人在学期间发表论文、出版专著、专利、获奖情况 (请在附件中附有关证明材料)					

发表论 文统计	国内刊物	国际刊物	国内学术 会议	国际学术 会议	其中发表 在核心期 刊	其中被 SCI （EI）收录 篇数
	1 篇	5 篇	3 篇	0 篇	6 篇	6 篇
第一 作者	1 篇	5 篇	3 篇	0 篇	6 篇	6 篇
论文名称		全部作者署名排序		发表时间		刊物名称
专著 统计	出版专著数量			出版专著数量（第一作者）		
	0 部			0 部		
专著名称		全部作者署名排序		出版时间		出版社名称
专利 统计	已授权发明或实用新型专利数			已授权发明或实用新型专利数 （第一发明人）		
	2 件			0 件		
专利号		专利名称		全部发明人排序		专利权人
获奖 统计	国家级奖励		省部级奖励			校级奖励
	1 项		0 项			3 项
奖励名称		奖励级别		全部获奖人排序		

三、本人开展的科研工作及取得的成果情况介绍

本人围绕国家双碳战略，通过设计和构筑新型金属氧化物/分子筛复合催化体系，实现 CO/CO₂ 加氢定向转化制备高价值甲醇、低碳醇等含氧化合物及烯烃、芳烃等烃类产品。同时，针对催化剂表界面结构的动态演变以及反应中间体的结构追溯，开展了相关机理研究，熟练掌握了原位红外、原位紫外、原位穆斯堡尔谱、飞行时间质谱及同位素标记示踪实验等的工作原理，并设计了相关实验方案，实现了 CO/CO₂ 加氢制烃和含氧化合物反应机理的准确解析。

目前，相关研究结果以第一作者在 Journal of the American Chemical Society、Applied Catalysis B、Science China Chemistry 等期刊发表论文 6 篇，其中 SCI 一区 top 期刊 4 篇；授权国家发明专利 2 项。荣获博士研究生国家奖学金、中国科学院大学“三好学生”、“三好学生标兵”等国家级、校级荣誉称号，具备扎实的科研能力和良好的团队协作精神。

四、在政治思想、道德品质、学风等方面自我评价

本人作为化学博士研究生，始终以严谨务实的作风要求自己，在政治思想、道德品质、学风建设等方面不断提升，努力成为兼具科研与综合素养的科研工作者，具体总结如下：

一、政治思想：坚定信念，筑牢思想根基

本人坚持正确政治方向，认真学习党的理论，关注国家科技政策，明确科技创新的重要意义，树立为国家化学科研事业贡献力量的信念。日常学习科研中，努力践行社会主义核心价值观，提升政治素养，抵制不良思潮，积极参与集体政治学习，将个人科研与国家需求相结合。

二、道德品质：严于律己，坚守为人底线

本人秉持诚信友善原则，恪守社会公德、职业道德和学术道德。与师长、同学相处中，尊重团结他人，分享科研经验，配合团队工作；科研中坚持实事求是，杜绝学术不端，严谨对待实验数据，注重个人品德修养，努力成为品德高尚的科研工作者。

三、学风：严谨务实，深耕科研之路

本人秉持“严谨、求实、创新、笃行”的学风，深耕化学研究。理论学习上，钻研专业理论与前沿成果，阅读中外文献，夯实基础、拓宽视野；实验中一丝不苟，规范操作、细致记录，积极解决实验问题，培养科学思维。科研中本人注重创新，探索新方向与方法，参与科研项目，坚持终身学习，虚心请教、参与学术交流，脚踏实地钻研，力争取得有价值的科研成果。

今后，我将继续提升综合素养，坚守信念、锤炼品德、深耕学术，务实进取，努力成为优秀的化学科研工作者，为国家科技发展贡献力量。

五、导师对申请人的评价及推荐意见

该生在CO₂催化转化方面取得重要进展，提出了低能耗中间
体CO₂加氢选择性制烯烃路线，低能耗收率达到创纪录
的36.6%。在JACS等期刊发表学术论文6篇，其中一区文章4篇，
同意并愿意推荐其申报中国科学院院士。

签名：



2026年 4月 9 日

五、研究所/院系推荐意见

负责人签名： (公 章)

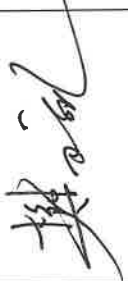
年 月 日

六、学校推荐意见

负责人签名： (公
章)

年 月 日

中国科学院院长奖学金申请者申请信息情况统计表

姓名	张倩	以第一作者发表论文情况（含学术会议）		SCI/EI 收录文章累计影响因子	参与专利	重要获奖荣誉
类别	博士	论文篇数	主要论文刊物名称（当年 IF）	左侧 IF 值相加值 71.4		
专业	化学		A、Journal of the American Chemical Society (15.6) (1 区) B、Applied Catalysis B: Environmental and Energy (21.1) (1 区) C、Applied Catalysis B: Environmental and Energy (21.1) (1 区) D、Science China Chemistry (9.8) (1 区) E、Frontiers in Chemistry (3.8) (3 区) F、燃料化学学报 (EI) G、中国化学会首届能源化学青年论坛，青岛，2022，会议摘要 H、第二十一届全国催化学术会议，昆明，2023，会议摘要 I、第二十届挥发性有机物污染防治化学年会，广州，2025，会议摘要			
联系电话	13831896923	6				
指导教师 审核签名						
发表论文题目列表:						
1、Enhancement of CO ₂ Hydrogenation to Light Olefins by Developing a Higher Alcohols-Intermediate Route (A)						
2、Hydrogenation of CO ₂ to higher alcohols on an efficient Cr-modified CuFe catalyst (B)						
3、Conversion of CO ₂ to higher alcohols on K-CuZnAl/Zr-CuFe composite (C)						
4、Recent advances in CO ₂ hydrogenation to higher alcohols (D)						
5、CO ₂ Hydrogenation on Metal-Organic Frameworks-Based Catalysts: A Mini Review (E)						
6、CO ₂ 加氢制备低碳烯烃的研究进展 (F)						
7、Cu/Uio-66 催化剂用于 CO ₂ 加氢制备甲醇 (G)						
8、In 改性 Cu-Fe 催化剂应用于 CO ₂ 加氢制低碳醇 (H)						
9、CO ₂ 加氢制低碳醇高性能 CuFe 基催化剂的研制 (I)						
发表专利:						
1、王森, 张倩, 樊卫斌, 董梅, 王建国: 一种双功能复合金属氧化物催化剂及其制备方法和应用, 已授权: ZL 2022 1 1458817.4						
2、王森, 张倩, 樊卫斌, 董梅, 王建国: 一种 Cr-K 改性铜铁复合氧化物及其制备方法和应用、二氧化碳加氢制低碳醇的方法, 已授权: ZL 2023 1 0307798.3						

附件 3

奖学金申请诚信承诺书

申请人：张倩，博士研究生，学号：202318004308004，身份证号：131182199410251621。本人已认真阅读《中国科学院山西煤炭化学研究所申报材料科研诚信提醒清单》（附件 4），对照提醒内容逐项自查确认。现郑重承诺：

在 2026 年度中国科学院院长奖学金申请中所使用材料、相关证明真实有效，无弄虚作假；所填报内容不存在科研失信行为。如有不实之处，自愿接受中国科学院大学和我所根据有关规定做出的处罚。

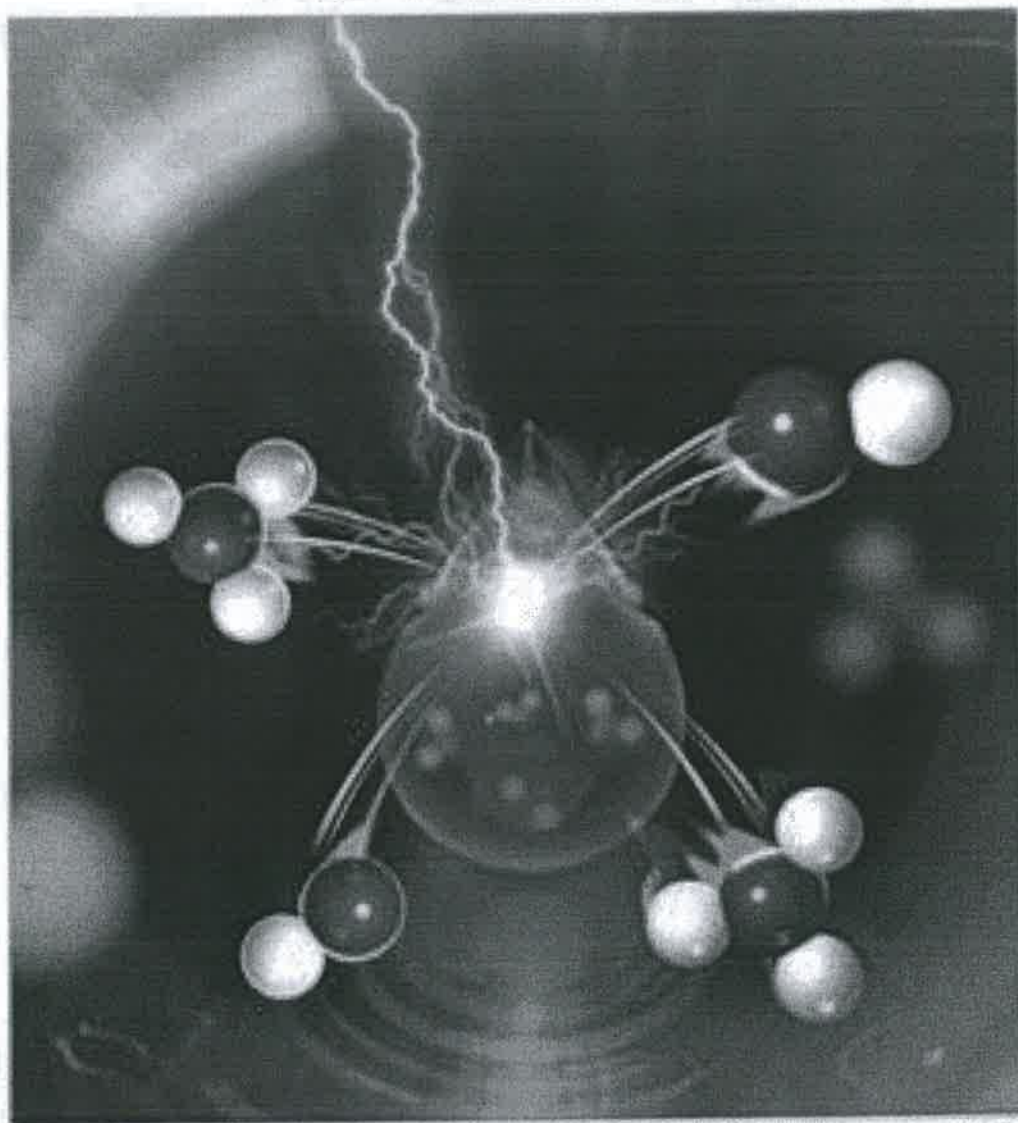
承诺人：张倩

日期：2026 年 4 月 8 日

December 10, 2023
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Enhancement of CO₂ Hydrogenation to Light Olefins by Developing a Higher Alcohols-Intermediate Route

Qian Zhang, Xiyu Li, Shifu Wang, Sen Wang,* Rui Geng, Pengfei Wang, Mei Dong, Jiangang Chen, Jianguo Wang, Xuning Li,* Unni Olsbye,* and Weibin Fan*

Cite This: *J. Am. Chem. Soc.* 2025, 147, 45070–45083

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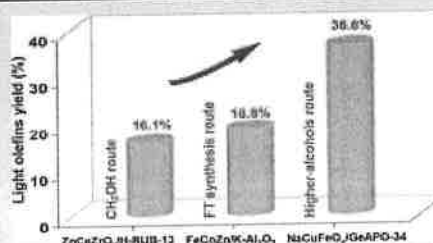
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ABSTRACT: CO₂ hydrogenation to light olefins generally occurs via Fischer–Tropsch (FT) synthesis and methanol-intermediate routes. However, these two routes usually give low light olefin yields, although large numbers of catalysts have been fabricated. This results from the limitation of the Anderson–Schulz–Flory (ASF) law or the formation of initial C–C bond and hydrocarbon pool (HCP) species. To overcome these problems, a higher alcohol-intermediate route is developed here as these alcoholic products are rapidly dehydrated to light olefins. The designed Na–CuFeO₄/H–GeAPO-34 composite shows CO₂ conversion of 75.1% and light olefins selectivity in all products (including CO) of 48.7%, thus resulting in an unprecedentedly high yield of 36.6%. In situ spectroscopy and DFT calculation results reveal that the metallic copper (Cu) species not only promotes iron (Fe) species reduction and carbonization by overflowing active hydrogen species and transferring electrons but also provides an effective site for stabilizing nondissociative CO* species. This enhances formation of CH₃* and nondissociative CO* species, which are coupled to generate large amounts of higher alcohols intermediates that are readily dehydrated into light olefins on weakly acidic H–GeAPO-34. This work confirms that the higher alcohols-intermediate route is highly effective for converting CO₂ into light olefins.



INTRODUCTION

Conversion of carbon dioxide (CO₂) into value-added fuels and commodity chemicals is an effective way to utilize CO₂ and achieve carbon recycling.^{1–7} Light olefins (C₂–C₄) as basic organic raw chemicals have been used for preparation of plastics, polymers, solvents, cosmetics, rubber, and pharmaceuticals.^{8–12} In 2024, global light olefin consumption reached ~800 million tons and its market scale reached around 400 billion dollars. Light olefins are generally obtained by steam cracking of naphtha and dehydrogenation of alkanes, but these processes require high energy input and high reaction temperature (≥550 °C), consequently consuming large amounts of energy and generating huge amounts of CO₂. In contrast, hydrogenation of anthropogenic CO₂ with renewable green hydrogen (H₂) into light olefins is an eco-friendly and sustainable carbon-neutral route, which, hence, attracts extensive attention.

Up to date, CO₂ to light olefins generally follows the Fischer–Tropsch (FT) synthesis or the methanol-intermediate routes.^{13–15} In FT synthesis route, CO₂ is first converted into CO through the reverse water–gas shift (RWGS) reaction and, then, undergoes dissociation and hydrogenation to form light olefins by coupling with CH_x species.^{16–18} Ma and co-workers reported a sodium- and manganese-modified Fe-based catalyst

(Fe–Mn–Na). It showed CO₂ conversion of 35.0% and light olefins selectivity of 28.1% in all products (including CO) at 340 °C and 2.0 MPa.¹⁹ The values were increased to 46.0 and 33.0%, respectively, on a honeycomb-structured graphene (HSG)-supported Fe–K catalyst (Fe–K/HSG) at similar conditions.²⁰ In the methanol-intermediate route, CO₂ is first adsorbed and activated on the surface oxygen vacancies of metal oxide component to form methanol intermediates, which then quickly diffuses onto the acid sites of zeolite and is converted into light olefins through a methanol-to-olefin (MTO) process.^{21,22} The mostly used metal oxides are Zn, Ga, and In-based oxides, and the generally employed zeolite component is H-SAPO-34 with 8-MR pore openings and CHA supercages.^{23–25} A typical catalyst is ZnZrO₄/H-SAPO-34 composite, which exhibits CO₂ conversion of 12.6% and light olefins selectivity of 80.0% in hydrocarbons + oxygenates (not including CO) at 380 °C and 2.0 MPa.²⁶ By replacing ZnZrO₄

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APPLIED C CATALYSIS B: ENVIRONMENTAL

Applied Catalysis B: Environmental
Volume 10, No. 1, 1991

Elsevier



Hydrogenation of CO₂ to higher alcohols on an efficient Cr-modified CuFe catalyst

Qian Zhang^{a,*,1}, Sen Wang^{a,*,1}, Rui Geng^{a,1}, Pengfei Wang^a, Mei Dong^a, Jianguo Wang^{a,1}, Weibin Pan^{a,1}

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ARTICLE INFO

Keywords:
 CO₂ hydrogenation
 Higher alcohol
 CuFe catalyst
 Cr modification
 Reaction mechanism

ABSTRACT

CO₂ hydrogenation to higher alcohols (C₂–OH) is a promising approach to achieve carbon recycling, but it is a challenge due to complex reaction network. Herein, various Cr-modified CuFe (Cr(x)-CuFe) catalysts were prepared, and their catalytic performance and reaction mechanism in CO₂ hydrogenation to C₂–OH were investigated. Introduction of small amounts of Cr enhances the interaction between Cu and Fe species, which alleviates CO over-dissociation and inhibits generation of iron carbides. In contrast, more acetate and acetaldehyde intermediates are produced via promoting the reaction of CH₃ with non-dissociated CO. Cr(1%)-CuFe showed CO₂ conversion, C₂–OH selectivity and space-time yield (STY) as high as 38.4%, and 29.2% and 104.1 mg g_{cat}⁻¹ h⁻¹, at 320 °C, 4.0 MPa and GHSV of 6000 ml g_{cat}⁻¹ h⁻¹. Interestingly, C₂–OH STY was further elevated to 265.5 mg g_{cat}⁻¹ h⁻¹, with a catalytic lifetime of at least 120 h, when GHSV was increased to 48000 ml g_{cat}⁻¹ h⁻¹.

1. Introduction

The combustion of large amounts of fossil fuels, e.g. coal and petroleum, causes excessive CO₂ emission into the atmosphere. In February 2023, the global CO₂ level in the atmosphere has reached 418.3 ppm [1,2], which leads to serious greenhouse effect, ocean acidification and other environmental problems [3,4]. Efficient conversion of CO₂ into high-value methane, gasoline, olefins, aromatics and oxygenates, etc. can not only reduce CO₂ emission and promote carbon neutrality, but also provide a new approach to produce fuels and commodity chemicals [5–10]. Higher alcohols (C₂–OH) are important building blockings, and have been widely used as eco-friendly additives, solvents and surfactants in modern chemical industry [11–13].

However, selective hydrogenation of CO₂ to higher alcohols (C₂–OH) is a challenge due to the high stability of CO₂ molecule and the complexity of C–C coupling reactions [14–18]. Llorca and co-workers anchored single Pd atoms onto Fe₂O₃ support, which showed an ethanol selectivity of 97.5%, along with the ethanol space time yield (STY) of 413 mmol_{ethanol} g_{cat}⁻¹ h⁻¹, at 300 °C and 0.1 MPa [19], although the CO₂ conversion is only 0.3%. They assumed that the interaction between Pd single atom and Fe₂O₃ support play a vital role in C–C coupling. A more active catalyst is metallic Pt nanoparticles (NPs)

supported on Co₃O₄ nanoplates (Pt/Co₃O₄-p) that gave a higher alcohols STY of 0.56 mmol g_{cat}⁻¹ h⁻¹ at 200 °C and 2.0 MPa. The synergic effect of Pt-Co nanoparticles and the surface oxygen vacancies of Co₃O_{4-x} promoted the activations of H₂ and CO₂, and thus, enhancing the formation of C₂–OH [20]. Recently, CeO₂-supported Pd dimer was found to exhibit an ethanol selectivity as high as 99.2% and a STY of 45.6 g_{ethanol} g_{cat}⁻¹ h⁻¹ at 240 °C, 3.0 MPa and 3000 ml g_{cat}⁻¹ h⁻¹ [21]. This is due to the unique Pd₂O₄ configuration that can directly dissociate CO₂ into CO and significantly promote the reaction of CH₃ and CO. Nevertheless, these noble metal-based catalysts are very expensive, limiting the large-scale applications.

In contrast, cheap and abundant CuFe-based catalysts receive a lot of attention in CO₂ hydrogenation to higher alcohols [22–29]. Sun and co-workers found that Na-modified CuFe catalyst prepared by the physical sputtering method showed high activity in CO₂ hydrogenation due to enhancement of CO₂ adsorption and Fe reduction and carbonization. The CO₂ conversion, ethanol selectivity and STY reached 32.3%, ~17% and 153 mg g_{cat}⁻¹ h⁻¹ at 310 °C, 3.0 MPa and 28,800 ml g_{cat}⁻¹ h⁻¹ [22]. At optimum condition, CuZnAl/K-CuMgZnFe bifunctional catalyst gave the C₂–OH selectivity and STY of 17.4% and 106.5 mg g_{cat}⁻¹ h⁻¹ [25]. Generally, higher alcohols are formed via the CO-intermediate route over CuFe-based catalysts. First, CO is generated through reverse

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APPLIED CATALYSIS B:

ENVIRONMENT AND ENERGY

Conversion of CO₂ to higher alcohols on K-CuZnAl/Zr-CuFe compositeQian Zhang^{a,b}, Sen Wang^{a,*}, Xuerong Shi^{c,*}, Mei Dong^a, Jiangang Chen^a, Juan Zhang^a,
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ABSTRACT

Direct conversion of CO₂ into higher alcohols (C₂+OH) is highly desirable, but rather challenging due to requiring the synergetic action of C-C coupling and CO insertion. Here, we developed a new K-CuZnAl/Zr-CuFe composite, which gave CO₂ conversion and C₂+OH selectivity of 40.6% and 22.4% respectively, while CO selectivity is only 10.3% at 320 °C, 4 MPa and 6000 mL g_{cat}⁻¹ h⁻¹. The C₂+OH STY can reach 195.1 mg g_{cat}⁻¹ h⁻¹, and is well maintained within 200 h at higher GHSV of 24000 mL g_{cat}⁻¹ h⁻¹. Introduction of K-CuZnAl and decrease of the contact distance of K-CuZnAl and Zr-CuFe boost the formation and subsequent conversion of CO* intermediate. In addition, doping small amounts of Zr into CuFe catalyst hinders the phase separation of Cu and Fe species by enhancing their interface interaction. As a result, the CH₃* species generated on iron carbide through CO* dissociative activation quickly reacts with the non-dissociative adsorbed CO* on adjacent Cu to produce more C₂+OH.

1. Introduction

Massive consumption of fossil fuels causes large amounts of CO₂ emission which is considered to cause a serious global greenhouse effect. However, on the other hand, CO₂ is a non-toxic and abundant carbon source [1,2]. By catalytic hydrogenation, it can be converted into light olefins, aromatics, liquefied petroleum gas (LPG), gasolines, etc [3–9]. This can not only effectively mitigate the CO₂ emission amount and alleviate the greenhouse effect, but also open up new ways to synthesize value-added chemicals.

Alcohols are important solvents, additives or essential building-block molecules for synthesis of esters, detergents and surfactants [10,11]. Nowadays, conversion of CO₂ into methanol has been industrialized. The mostly used catalysts are modified copper-zinc-aluminum (CuZnAl), although indium oxide (In₂O₃), zinc-zirconium solid solution (ZnZrO₄) and molybdenum sulfide (MoS₂) have been developed [12–15]. Compared with methanol, C₂+ higher alcohols (C₂+OH) with higher energy density are more difficult to be synthesized, due to requirement of synergetic execution of C-C coupling and CO insertion [16].

Pt, Pd, Rh or Ir-based noble metal catalysts have been proved to show excellent performance in CO₂ hydrogenation to C₂+OH [17–19]. Wang and co-workers reported that Na-Rh embedded in silicalite-1 catalyst

(Na-Rh@S-1) exhibited C₂+OH selectivity and space time yield (STY) of ~30% (ethanol accounting for 80% of C₂+OH) and 0.49 mmol g_{cat}⁻¹ h⁻¹ at CO₂ conversion of 10% [20]. The space confinement effect of silicalite-1 endows high catalytic stability with the catalytic life of 100 h. By optimizing Pt loading and reaction conditions, ordered Pt/CeO₂ sample gave the ethanol selectivity and STY of 82.0% and 0.51 mmol g_{cat}⁻¹ h⁻¹ [21]. Recently, a more interesting single-atom catalyst of Ir-In₂O₃ was developed by Ye and co-workers. This catalyst showed the ethanol selectivity and STY as high as 99% and 0.99 mmol g_{cat}⁻¹ h⁻¹ [22], as the isolated Ir atom anchored on the oxygen vacancies of In₂O₃ formed Lewis acid-base pair, which not only promotes activation of CO₂ to CO*, but also accelerates the C-C coupling between CO* and methoxy group. Regardless of the good catalytic results of these noble metal-based samples, their high prices limit the large-scale industrial applications.

Thus, design of CuFe-based catalysts has attracted great interest due to their good performance and low price [23]. Liu et al. found that Cs-CuFeZn catalyst was a potential candidate for the formation of higher alcohols, with the selectivity and STY of 19.8% and 73.4 mg g_{cat}⁻¹ h⁻¹, along with CO₂ conversion of 36.6%, at 330 °C and 5.0 MPa [24]. The selectivity of C₂+OH can be further elevated to 22.8% on K-CuZnMgFe catalyst, despite that its STY is slightly decreased to 69.6 mg g_{cat}⁻¹ h⁻¹ due to the decline of CO₂ conversion [25]. For CuFe-based catalysts, it is

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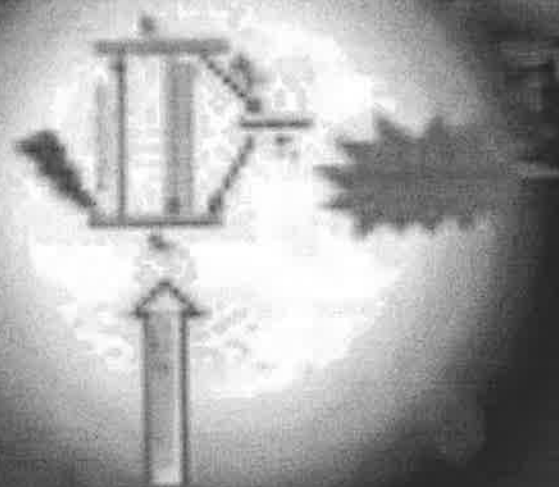
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Recent advances in CO₂ hydrogenation to higher alcoholsQian Zhang^{1,2}, Sen Wang^{1*}, Mei Dong¹, Jianguo Wang^{1,2} & Weibin Fan^{1*}¹State Key Laboratory of Coal Conversion, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan 030001, China;²University of Chinese Academy of Sciences, Beijing 100049, China

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CO₂ hydrogenation to higher alcohols (C₂–OH) is an effective way to realize carbon recycling, which can not only reduce the CO₂ amounts in atmosphere and mitigate the greenhouse effect, but also provides a new route to synthesize important chemicals. However, this process is a challenge because the inert CO₂ molecule is difficult to be activated and undergo C–C coupling. The key to achieve selective conversion of CO₂ to C₂–OH is to design high-performance catalytic systems and unravel the reaction mechanism. In this review, we report several typical CO₂ hydrogenation-to-C₂–OH catalyst materials, including noble-metal catalysts, Cu-based catalysts, Co-based catalysts and Mo-based catalysts, and evaluate the effects of various promoters on the catalytic performance and reaction mechanism. It will provide not only fundamental insights into the CO₂ hydrogenation-to-C₂–OH reaction mechanism, but also guidance for the development of related high-performance catalysts.

CO₂ hydrogenation, higher alcohols, catalysts, promoters, reaction mechanism

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1 Introduction

The extensive use of fossil fuels such as coal, petroleum and natural gas, has promoted the rapid development of human society, but it has also led to a large amount of CO₂ emissions. Over the past 200 years, the CO₂ concentration in the atmosphere has increased from 270 ppm to 423 ppm, along with the elevation of global temperature by 1.2 °C. It results in serious greenhouse effect and a series of environmental problems, such as climate warming, ocean acidification and sea level rise. [1]. Therefore, decreasing CO₂ emissions to atmosphere has become a common challenge in the whole human society. Nowadays, carbon capture, utilization and storage (CCUS) are considered as an effective approach to achieve carbon neutrality. CO₂ utilization can not only mitigate CO₂ emission and reduce the use of fossil fuels but also

bring up a sustainable CO₂ cycle-economy.

CO₂ is a highly stable molecule ($\Delta G_f^\circ = -394.4 \text{ kJ mol}^{-1}$). Its C=O bond dissociation energy reaches $\sim 800 \text{ kJ mol}^{-1}$, being much higher than those of C–C (336 kJ mol^{−1}), C–O (327 kJ mol^{−1}) and C–H (411 kJ mol^{−1}) bonds [2]. This means that the CO₂ activation requires the input of substantial energy or using a high-energy density reductants (e.g., hydrogen (H₂)) to break its stable C=O bond [3,4]. By catalytic hydrogenation, CO₂ can be effectively converted into value-added hydrocarbons and oxygenates such as methane [5,6], olefins [7–9], aromatics [10,11], liquefied petroleum gas [12] and alcohols [13–15] (Figure 1). Higher alcohols (C₂–OH) are important eco-friendly additives, solvents and potential hydrogen carrier. In addition, these alcohols can be directly transformed into olefins and surfactants [16].

Higher alcohols are generally obtained through the fermentation of carbon-based agricultural raw materials (corn and sugar cane) and petroleum-derived olefin hydration [17].

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CO₂ Hydrogenation on Metal-Organic Frameworks-Based Catalysts: A Mini Review

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Conversion of carbon dioxide (CO₂) into value-added fuels and chemicals can not only reduce the emission amount of CO₂ in the atmosphere and alleviate the greenhouse effect but also realize carbon recycling. Through hydrogenation with renewable hydrogen (H₂), CO₂ can be transformed into various hydrocarbons and oxygenates, including methanol, ethanol, methane and light olefins, etc. Recently, metal-organic frameworks (MOFs) have attracted extensive attention in the fields of adsorption, gas separation, and catalysis due to their high surface area, abundant metal sites, and tunable metal-support interface interaction. In CO₂ hydrogenation, MOFs are regarded as important supports or sacrificed precursors for the preparation of high-efficient catalysts, which can uniformly disperse metal nanoparticles (NPs) and enhance the interaction between metal and support to prevent sintering and aggregation of active metal species. This work summarizes the recent progress on hydrogenation of CO₂ to methanol, methane and other C₂₊ products over various MOFs-based catalysts, and it will provide some clues for the design of MOFs materials in energy-efficient conversion and utilization.

Keywords: MOFs, CO₂ hydrogenation, methanol, methane, C₂₊ Products

INTRODUCTION

Due to the rapid consumption of fossil resources, e.g., coal, petroleum, and natural gas, a large number of CO₂ have been released into the atmosphere (Song, 2006). From 2006 to 2021, the global CO₂ concentration in the atmosphere has been elevated from 381 to 415 ppm (NOAA, 2022). The massive emission of CO₂ has brought serious environmental problems, such as global climate change and ocean acidification (Valles-Regino et al., 2015). Hence, reduction of CO₂ amount and mitigation of greenhouse effect are the major challenges faced by the whole human society.

Regardless of this, CO₂ is an important C1 platform molecule. Conversion of CO₂ through sustainable catalytic processes into valuable chemicals and clean fuels is a promising way for CO₂ utilization which could promote a circular carbon economy (Srinivas et al., 2014; Dudas et al., 2015; Porosoff et al., 2016; Rafiee et al., 2018). CO₂ conversion can be achieved by electro-catalysis, photo-catalysis, and thermal-catalysis processes. Electro-catalysis or photo-catalysis from clean and renewable electrical or solar energy is regarded as an important route for CO₂ reduction reaction (CO₂ RR). Through the rational design of high efficient catalysts, these reactions can be performed under relatively mild conditions that considerably decrease the energy consumption (Liu et al., 2012; Handoko et al., 2013; Jhong et al., 2013; Wang et al., 2015; Perathoner and Centi, 2019; Wang J.-J. et al., 2021; Zhang et al., 2021; Zhang et al., 2022a). Nevertheless, the electro-catalysis or photo-catalysis for CO₂ conversion is time or geographically dependent, which, thus, decreases their



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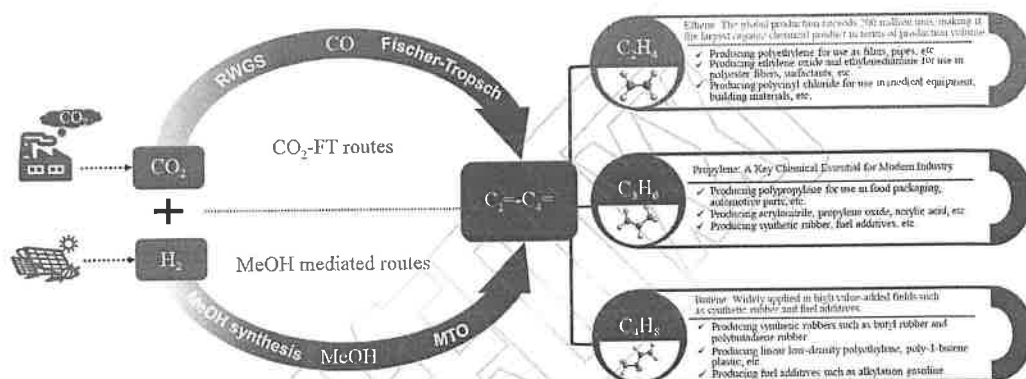
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CO₂ 加氢制备低碳烯烃的研究进展

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摘要: CO₂ 加氢制低碳烯烃过程的成功开发和应用不仅可以减缓因温室气体大量排放而带来的环境问题, 同时也开辟了制备重要化学品的新路线。由于 CO₂ 分子高度稳定, 实现其高



效且定向活化和转化极具挑战。本文综述了当前 CO₂ 加氢制低碳烯烃的研究进展, 重点分析了费托合成(CO₂-FTO)路线和甲醇中间体(CO₂-MTO)路线的优缺点。介绍了影响 CO₂ 活化和低碳烯烃生成的关键因素, 包括助剂种类、载体结构以及表面缺陷位点浓度等。同时, 研究了分子筛孔道结构和酸性对甲醇转化制烯烃的影响机制。

关键词: CO₂ 加氢; 低碳烯烃; 费托合成路线; 甲醇中间体路线

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Recent advances in CO₂ hydrogenation to light olefins

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Abstract: The successful development and application of CO₂ hydrogenation to light olefins can not only alleviate the environmental issues caused by the massive emission of greenhouse gases, but also open up a new way for the preparation of important high-value chemicals. Due to the chemical inertness of CO₂ molecules, selectively activation and conversion of CO₂ into target light olefins remain highly challenging. In this work, we review the current research progress in the hydrogenation of CO₂ to light olefins, with a focus on analyzing the advantages and disadvantages of the Fischer-Tropsch synthesis (CO₂-FTO) route and the methanol intermediate (CO₂-MTO) route. The key factors, such as the type of additives, the structure of the carrier and the concentration of surface defect sites, affecting the activation of CO₂ and the formation of light olefins are discussed in detail. Meanwhile, essential role of the pore structure and acidity of zeolites in conversion of methanol intermediates to olefins is analyzed.

Key words: CO₂ hydrogenation; light olefins; Fischer-Tropsch synthesis route; methanol intermediate route

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Cu/UIO-66 催化 CO₂ 加氢制甲醇[†]张倩^{1,2}, 王森^{1,*}¹ 中国科学院山西煤炭化学研究所, 太原, 030000² 中国科学院大学, 北京, 100049

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二氧化碳(CO₂)转化制备高附加值化学品不仅可以缓解由于CO₂大量释放带来的温室效应,同时也开辟了一条制备清洁燃料的新途径。甲醇是现代化学工业中重要的基础原料;通过加氢,可将CO₂直接转化为高附加值的甲醇。金属有机框架(MOFs)是一类以金属-氧簇作为次级构筑单元并与有机配体相连接所构筑的多孔固体材料。MOFs具有高比表面积、结构可调等优势^[1]。例如,以UiO-66作为载体可以提高金属Cu分散度,增强金属Cu和载体之间的相互作用进而防止Cu聚集失活,因而在CO₂加氢制甲醇中广泛应用。最近的研究表明,孤立的Cu颗粒加速了逆水煤气反应生成,促使CO₂转化生成CO;而当Cu负载到UiO-66上,会使Cu和Zr₆O₈相互作用形成Cu-O-Zr界面有利于甲醇生成^[2]。基于此,本文研制了一系列具有不同Cu负载量的Cu/UIO-66催化剂,并探索其在CO₂加氢制备甲醇中的催化性能和反应机理。结果表明,当Cu负载量为1.3wt%时,甲醇的时空产率高达35mmol/g_{Cu}/h;而Cu负载量由1.3wt%提高到5wt%时,甲醇的选择性由45%提高到54%,说明较高Cu负载量有利于生成更多Cu-O-Zr活性位点,促进甲醇生成。针对反应后的催化剂进行结构表征发现,反应之后UiO-66的XRD衍射峰依然较强,说明UiO-66结构没有被破坏,表现出较高的稳定性。

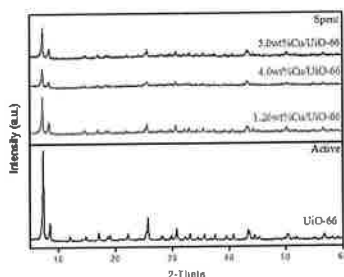


Fig. 1 XRD patterns of the spent catalysts

关键词: Cu/UIO-66, CO₂ 加氢, 甲醇

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In 改性 Cu-Fe 催化剂应用于 CO₂ 加氢制低碳醇*张倩^{ab}, 王森^{a*}, 董梅^a, 樊卫斌^{a*}^a 中国科学院山西煤炭化学研究所, 太原, 030000^b 中国科学院大学, 北京, 100049

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关键词: CO₂ 加氢, 低碳醇, Cu-Fe 催化剂, In 助剂

CO₂ 加氢高效催化剂的研发和应用不仅可以缓解因 CO₂ 大量排放导致的温室效应, 同时也开辟了制备高附加值大宗化学品的新路线。乙醇等低碳醇 (C₂+OH) 具有较高的能量密度, 在现代化学工业中具有广泛的市场需求。CuFe 催化剂在 CO₂ 加氢制低碳醇过程中应用广泛。在 CuFe 催化剂中, CO₂ 首先经过逆水煤气反应生成 CO, 产生的 CO 在碳化铁 (如: Fe₅C₂) 上进行解离生成 CH_x, 并与相邻 Cu 位点上的非解离吸附的 CO 进行 C-C 偶联生成乙醇^[1]。但是, 由于 C-C 耦合过程的高能垒以及反应过程的复杂性, 导致直接转化 CO₂ 制乙醇等低碳醇非常困难。In 作为结构助剂可以提高催化剂表面的氧空穴浓度, 促进 CO₂ 吸附和活化^[2]。结果表明, 在 CuFe 催化剂中引入 In 助剂, CO 和低碳醇的选择性均明显提高, 证实了 In 存在促进逆水煤气反应, 有利于 CO 中间体生成, 并进而有助于 CO 与 CH_x 进行 C-C 偶联生成低碳醇^[3], 使得低碳醇的选择性从 18.9% 提高到 24.1%。XRD 结果表明, 只能观察到 CuO 的衍射峰, 没有观察到 In₂O₃ 和 Fe₂O₃ 衍射峰, 说明 In₂O₃ 和 Fe₂O₃ 两者分布均匀。

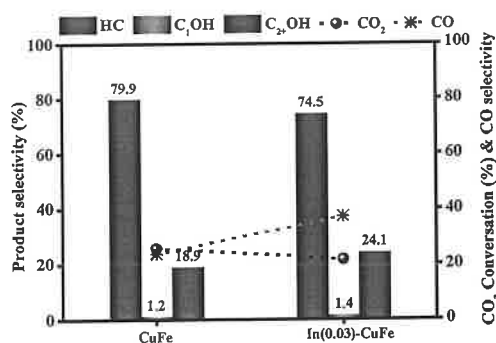


Fig.1 Catalytic performance

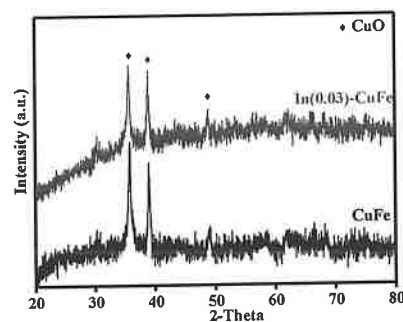


Fig. 2 XRD patterns of the fresh catalysts

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CO₂ 加氢制低碳醇高性能 CuFe 基催化剂的研制

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CO₂ 加氢高效催化剂的研发和应用不仅可以缓解因 CO₂ 大量排放导致的温室效应，同时也开辟了制备高附加值大宗化学品的新路线。乙醇等低碳醇具有较高的能量密度，在现代化学工业中具有广泛的市场需求。CuFe 催化剂被广泛用于 CO₂ 加氢制低碳醇。在该催化体系中，CO₂ 首先经过逆水煤气反应生成 CO，并在碳化铁（如：Fe₅C₂）上进行解离生成 CH_x；这些 CH_x 随后与相邻 Cu 位点上非解离吸附的 CO 进行 C-C 偶联生成乙醇^[1,2]。

对于铜铁催化体系, 增强金属铜和碳化铁界面相互作用, 有助于提高 $\text{C}=\text{O}$ 键活化和 CO^* 插入能力, 进而促进低碳醇的生成。研究发现, 在 CuFe 催化剂中引入适宜的 Cr 助剂, 促进了 Cu-FeC_x 界面的生成, 有利于低碳醇的产生。与 CuFe 催化剂相比, 引入 $\text{Cr}(1\text{wt.}\%)$ 助剂后, CO_2 转化率从 25.5% 提升至 33.5%, CO 选择性从 23.6% 降低至 16.0%, 低碳醇选择性从 18.9% 提升至 24.9%, 低碳醇时空产率从 39.2 提升到 $76.3\text{ mg g}_{\text{cat}}^{-1}\text{ h}^{-1}$ (反应条件: $300\text{ }^\circ\text{C}$, 4 Mpa , 空速 $6000\text{ mL g}_{\text{cat}}^{-1}\text{ h}^{-1}$) (图 1a)。通过优化反应条件, 低碳醇的选择性和时空产率可进一步提升至 29.2% 和 $104.1\text{ mg g}_{\text{cat}}^{-1}\text{ h}^{-1}$; 低碳醇中乙醇占比达 80%, 稳定性可维持 200h 以上 (图 1b)。结合原位光谱、同位素标记实验和理论计算结果证实, Cr 助剂的存在诱导了电子由 Cu 向 Fe 转移, 加强了 Cu-Fe 之间的界面相互作用 (图 1c)。富电子的铁位点更容易碳化生成碳化铁物质, 促进了 CO 解离形成 CH_x^* ; 同时, 金属 Cu 位点稳定了部分非解离吸附的 CO^* 。 CH_x^* 与 CO^* 通过快速插入生成了乙酸盐、乙醛等中间体, 并进一步加氢形成乙醇等低碳醇。

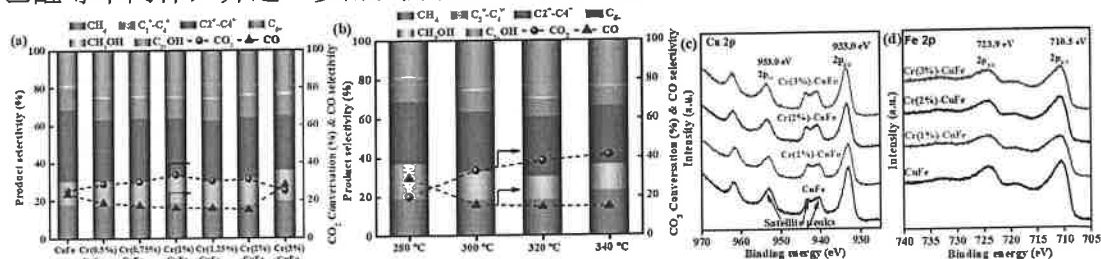


图 1. (a) CuFe 和不同 Cr 含量的 Cr-CuFe 催化剂的催化活性(反应条件: 300 °C, 4Mpa, 空速 6000 mL g_{cat}⁻¹ h⁻¹); (b) 不同反应温度 Cr(1wt.%) -CuFe 催化剂性能测试; (c) Cr-CuFe 催化剂的 Cu (2p)XPS 和(d) Fe (2p)XPS。

关键词: CO₂ 加氢、低碳醇、Cr 改性 CuFe 催化剂

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发明专利证书

发明名称：一种Cr-K改性铜铁复合氧化物及其制备方法和应用、二氧化碳加氢制低碳醇的方法

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