

基于离子液低共熔溶液的硫碳协同资源化工艺的构建与发展

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在“双碳”目标的指引下, CO₂减排已成为国际社会共同关注的焦点。当前, 碳捕获、利用与封存(CCUS)技术中的CO₂捕集是减少CO₂排放的主要手段。而同为酸性气的H₂S常与CO₂共存, 由于二者在醇胺溶液中的吸收动力学存在显著差异, 常用超重力技术实现CO₂和H₂S的分离与捕集^[1]。H₂S可通过湿法氧化法直接氧化为硫磺, 但如何将捕集的巨量CO₂转化为可资源化的化学品是实现“双碳”目标的关键。

CO₂资源化途径很多, 比如通过加氢、光化学以及电化学等还原技术将其转化为合成气(CO + H₂)、CH₄以及低碳有机化合物等, 这些过程均需要氢源与活化的CO₂分子发生反应才可以得到产物, 其中氢源来自反应体系中的H⁺、介质H₂O或反应生成的H原子及外源提供的H₂。

湿法氧化H₂S在生成硫磺的同时脱硫富液中产生游离的H⁺, 采用电解法再生脱硫富液时游离H⁺可在电极表面形成活泼的H原子。无论H⁺还是H原子都是CO₂加氢还原不可或缺的基础原料, 因此, 以H₂S为氢源发展功能化溶液体系, 将H₂S的湿法氧化脱硫与电化学还原CO₂相结合, 可以构建硫碳协同资源化的新工艺(carbon & sulfur capture and utilization, CSCU)。据不完全统计, 我国每年H₂S排放量约为80亿m³^[2], CO₂排放量约580余亿m³, 将H₂S氧化与CO₂的资源化相结合, 在减排20~80亿m³的CO₂同时得到大量的可燃气体(CO、CH₄)或化工产品(HCOOH、CH₃OH), 由此可见, CSCU工艺在建设绿色化学工厂, 实现“双碳”目标上, 具有重大的研发意义和应用潜力。

1 水溶液中硫碳协同反应

硫碳协同反应的本质是利用H₂S作为氢源, 电化学还原H⁺生成活性原子H, 实现对CO₂的加氢还原^[3]。Li及合作者^[4]报道了水溶液间接电解H₂S与电还原CO₂的联合工艺, 即用整合铁([Fe(III)EDTA]⁻/[Fe(II)EDTA]²⁻)氧化H₂S, 然后电还原脱硫富液中H⁺, 并将其对CO₂加氢得到CO。因此, 通过间



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接电解工艺在强化H₂S氧化过程硫磺分离的基础上, 实现电化学还原溶液中H⁺并对CO₂加氢处理。

为了获得长效脱硫运行效果, 整合铁水溶液的pH一般控制在8~9, 因此H₂S被氧化后释放的H⁺被碱性溶液中和。Wang等人^[2]报道了强化间接电解制氢的电子介导-离场电催化技术: Fe(III)首先与H₂S反应生成硫磺和Fe(II), Fe(II)在阳极区被氧化再生, H⁺则迁移至阴极区, 在V(II)的作用下催化还原成H₂, V(II)则被氧化为V(III), 最后V(III)在阴极又被还原成V(II)而实现再生。该过程中Fe(III)/Fe(II)与V(III)/V(II)作为两对氧化还原介体加速了H₂S的全电解, 并避免了硫磺对反应过程的干扰。

氢氧化钠、碳酸钠等碱性溶液通常被用于捕集CO₂气体, 电化学还原反应中的氢源主要由H₂O提供, 所得CO₂电催化还原产物主要包括甲烷和甲醇两大类。CO₂的甲烷化反应主要包括中间体路径和缔合反应路径^[5], 前者是CO₂先被还原成CO再加氢得到CH₄, 而后者则被认为CO₂可以不生成CO中间体, 直接通过CO₂→HCOO*→HCO*→CH_x→CH₄的路径得到甲烷。而CO₂生成甲醇的路径则更为复杂, 胡博等人^[6]将其归纳为四种反应途径, 分别为r-HCOO*机理、HCOO*机理、RWGS+CO-Hydro机理(RWGS: 逆水煤气反应)与反式

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COOH机理。

虽然CSCU工艺是一种清洁、绿色的硫碳资源化技术,然而水相体系中存在诸多局限性。对于CO₂电化学还原过程,电解液的特性直接影响到CO₂电化学还原产物的选择性^[7]。Koper团队^[8]的研究表明,不同的CO₂还原路径对pH的依赖性存在着明显差异。此外,电解液中阳离子的类型对产物的选择性具备较大的影响,Murata等人^[9]的结果表明,随着阳离子水合物半径的增加(Li⁺到Cs⁺),外亥姆霍兹平面(OHP)电位发生变化,从而影响电极界面上的质子浓度,进而影响烃类选择性和反应速率。除阳离子外,阴离子同样能够影响CO₂的电催化还原,研究发现卤离子类型(Cl⁻、Br⁻、I⁻)能够影响CO₂加氢还原产物选择性,由于Cl⁻和Br⁻能够吸附在电极界面上增加界面负电荷,从而有利于CO生成^[10]。

此外,由于CO₂还原是一个多步骤的复杂反应,产物的选择性与电极材料属性密切相关,难以实现甲烷或甲醇产物的定向转化。并且,该技术面临从水溶液中分离复杂CO₂还原产物的挑战。

2 有机介质中硫碳协同反应

间接电解H₂S制氢需要将捕集的H₂S定向转化为硫磺并将其从反应体系中分离。Hua等人^[11]提出了非水相湿法氧化脱硫方法,将三氯化铁溶解于强极性非质子溶剂中,无需调节溶液的酸碱性,实现对H₂S的吸收与转化。H₂S被氧化后生成的H⁺通常会以氢键的方式与溶剂分子结合,生成对应的阳离子,而阳离子又倾向于向阴极扩散,从而为CO₂的电还原过程提供质子。有机溶剂与氢离子的结合主要依托于氢键弱相互作用,相对于水解反应,更加有利于H⁺的释放,从而提高CO₂加氢效率。

有机体系电化学还原CO₂有两点优势:(1)存在强氢键网络。游离态H⁺容易与有机分子中的氢键供体结合,从而限制H⁺的迁移,能够有效抑制HER反应,提升CO₂电化学还原产物的选择性;(2)CO₂在某些有机溶剂中溶解度远高于水溶液中,如室温下CO₂在甲醇中的溶解度是其在在水中溶解度的5倍^[12]。Moret等人^[13]对比水和DMSO对CO₂加氢制备甲酸的性能发现,相同条件下水溶液中仅能形成0.2 mol/L甲酸,而在DMSO中甲酸的生成量可高达1.9 mol/L。Huang等人^[14]报道,在80 mmol/L氢氧化钠甲醇溶液中CO₂电还原制甲烷的法拉第效率可达到62%。尽管有机体系能够提高产物的选择性,但有机介质的导电性和稳定性较差,电催化氧化导致的分解难题需要解决。

3 离子液低共熔溶液中硫碳协同反应工艺

3.1 铁基离子液低共熔溶液的物化特性

铁基离子液体是由螯合铁阴离子([FeX₄]⁻、[FeEDTA]⁻)和阳离子组成的室温熔融盐液体,既具有离子液体的导电

性、饱和蒸气压低、电化学稳定性高等特性,还具有氧化性和Lewis酸催化活性等优势^[15-17]。低共熔溶剂是由一定化学计量比的氢键供体和氢键受体组合而成的多组分低共熔混合物,其中非质子溶剂就是常用的组分。离子液体的阴阳离子与非质子溶剂分子间通过氢键作用促进金属基离子液体溶于低共熔溶剂中,形成新型离子液低共熔溶液^[18](MIL-DES)。由于溶剂化效应,金属基离子液中离子对的相互作用被低共熔溶剂显著弱化,离子液低共熔溶液(MIL-DES)中充斥着相对自由的阴阳离子或离子对,溶液黏度降低,提高液相传质效率,这类材料相较于传统离子液体更容易实现功能化调控,拓宽其应用潜力。

研究表明,由BmimFeCl₄、NHD(聚乙二醇二甲醚)、MDEA(*N*-甲基二乙醇胺)组成的三元铁基离子液低共熔溶液在吸收氧化H₂S过程中释放出的H⁺与MDEA结合形成亲水性更强的[MDEAH]⁺相,富集H⁺和亚铁络合阴离子;而剩余的BmimFeCl₄留存于NHD相中而形成液液两相体系^[19],此相变形成原理与相变溶剂吸收CO₂一致。

3.2 铁基离子液低共熔溶液的间接电解脱硫

余江课题组^[16]于2010年提出基于铁基离子液体的非水相湿法氧化脱硫新工艺(Nasil),使用BmimFeCl₄定向氧化H₂S为硫磺,无副盐生成,硫磺易分离且可直接熔硫纯化得到高纯度硫磺,而脱硫富液可通过鼓入空气或电解实现再生^[20]。然而,BmimFeCl₄的强Brønsted酸性不利于其对H₂S、CO₂等酸性气体的吸收,同时四面体结构[FeCl₄]⁻的空间位阻效应显著地抑制了铁离子的氧化活性,不利于H₂S的定向转化^[21],未转化的H₂S会严重干扰电解过程中电极表面反应。与非质子溶剂复配后,铁基离子液低共熔溶液的气液传质性能和铁离子的氧化活性均得到显著改善^[22,23]。Guo等人^[24]构建的Fe(Ⅲ/Ⅱ)-IL/DMF脱硫体系,Fe(Ⅲ)先与H₂S反应生成Fe(Ⅱ)和H⁺,分离硫磺后再对脱硫富液进行电解再生,H₂S中的H⁺在阴极被还原成H₂,而Fe(Ⅱ)在阳极则被氧化为Fe(Ⅲ)。但是,脱硫富液黏度高和H⁺浓度低导致电解液中传质效率和法拉第效率均较低,需要有针对性地解决。

由BmimFeCl₄、NHD、MDEA组成的三元铁基离子液低共熔溶液脱硫相变发生过程,不仅增加了[MDEAH]⁺相中H⁺的浓度,还可显著减少电解再生脱硫富液的体积,利于电解效率的提高。[MDEAH]⁺相的强极性亲水特性能够抑制疏水硫磺分子与溶剂分子的作用,提高硫磺分离效率。相变过程首先发生在硫磺吸收阶段,CO₂气体分子和H₂S被氧化后释放出的大量H⁺与醇胺分子相互作用,导致复合体系各组分疏水性发生变化而分相,进而对特定组分起到富集的作用。

3.3 铁基离子液低共熔溶液中CO₂活化及电催化还原

非水电解液电还原CO₂过程中阳离子活化效果很大程度

上取决于氢键供体的强度,添加氢键供体能够促进质子与电子的转移.研究表明,离子液体是最具有潜质的 CO_2 电化学还原电解质之一,离子液体内部的氢键网络能够束缚游离态的 H^+ ,有效抑制HER反应发生,提高 CO_2 电还原反应选择性^[25].咪唑环上的C4-H和C5-H能够与 CO_2 形成氢键,从而导致 CO_2 的键角弯曲而被活化^[26], CO_2 可通过咪唑环上的C2形成Im- CO_2 加合物以强化 CO_2 RR反应^[27].阴离子也能够影响 CO_2 RR过程的反应活性与选择性,如BmimNTF₂具备最大的甲酸盐法拉第效率(38.7%),而BmimDCA表现出了最高的制氢选择性^[28].

3.4 铁基离子液低共熔溶液的硫碳协同反应工艺构建

综上所述,本文提出一种基于 H_2S 和 CO_2 的联合脱硫脱碳资源化新工艺(CSCU)设想,如图1所示:以铁基离子液低共熔溶液为脱硫液吸收酸性气 H_2S 和 CO_2 , H_2S 首先被 Fe(III) 氧化为硫磺并分离(A).随着脱硫液中 H^+ 和 CO_2 含量增加,脱硫富液自发形成富酸相和贫酸相的双相体系(B),其中 Fe(II) 、 H^+ 和 CO_2 富集于富酸相(C),进入电解槽成为待再生的新脱硫富液(D).富酸相中 CO_2 被活化后与阴极电极表面产生的活性 H 原子发生氢化反应生成目标化学品(E),而阳极电极表面发生 Fe(II) 的氧化再生转化为 Fe(III) (F),富酸相再生后经过逆相变所得NHD相与贫酸相混合后循环使用(G).

一般情况下,以富酸相为电解液的电解过程中, Fe(III)/Fe(II) 和 H^+/H_2 在阴阳极电极表面的竞争反应将导致电解过程难以控制.但是,在电解富酸相脱硫富液的初始阶段,高浓度的 Fe(II) 和 H^+ 使得阴极发生 H^+/H_2 还原制氢反应和阳极发生 Fe(III)/Fe(II) 的氧化再生反应,随富酸相的酸度及极性的降低,

溶于富酸相中的弱极性NHD相开始析出,发生逆相变过程,此时再生得到的 Fe(III) 被萃取转移到贫酸NHD相中,及时消除了 Fe(III) 在阴极电极表面与 H^+ 的电还原竞争反应(图1).电解过程中的逆相变萃取原理为构建无隔膜电解再生脱硫富液工艺奠定了基础.

3.5 离子液低共熔溶液CSCU工艺面临的挑战

理论上构建的联合脱硫脱碳新工艺仍面临实际体系复杂性的挑战.

(1) 原料气工况复杂性.原料气组成除了 H_2S 、 CO_2 、 CH_4 ,还含有低碳烃和水,而不同类型原料气在温度、压力、浓度等工况条件的差异对脱硫体系及工艺的构建有重大影响.如天然气中的有机硫和水被溶液吸收后在电解过程可能引发较为复杂的副反应.水作为非质子溶剂,对低共熔溶剂分子间氢键网络结构有明显影响,能够降低黏度,提高气液传质性能和电解液的电导率,从而增加电流密度和产氢量.然而,这也可能导致HER反应增加,不利于 CO_2 还原.

(2) 溶液体系稳定性.反应过程中所产生的硫磺需要在 $130\sim 150^\circ\text{C}$ 通过熔硫的方式分离提纯,分离后的脱硫液将被循环使用,因此需要脱硫液体系具有很好的热稳定性;此外,在电解再生脱硫液的过程中,应尽可能减少副反应,这要求脱硫体系具有良好的化学稳定性,以避免脱硫液组分流失或降解.

(3) 目标产物选择性.CSCU工艺的产物主要包括硫磺、合成气或甲烷/低碳醇/低碳含氧酸.对硫磺而言,熔硫过程中液硫与脱硫液需要高效分离以保障硫磺的纯度.至于 CO_2 的资源化,需要考虑原料气组成、温度和压力等工况差异,强化电催化还原过程控制,并设计目标反应路径以得到定向产

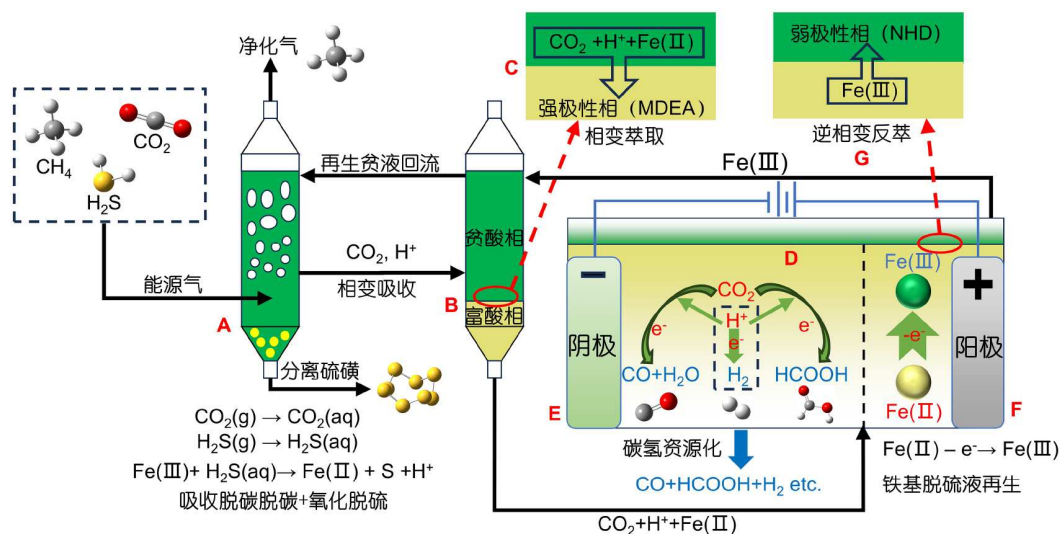


图1 (网络版彩色)铁基离子液低共熔溶液的硫化氢-二氧化碳协同资源化工艺示意图

Figure 1 (Color online) Schematic diagram of hydrogen sulfide-carbon dioxide synergistic resource recovery process for iron-based ionic liquid deep eutectic solution

物。如焦炉煤气的CO₂可以转化为合成气(CO + H₂),而天然气的净化最优产物则为甲烷,在石油化工生产过程中,则可选择甲酸、甲醇等化工产品作为产物。

(4) 工艺集成及过程控制。为保证高效电解过程,吸收剂通常需要维持较高的酸度,但相变吸收剂酸度的提升可能会导致黏度显著增加,从而降低气液传质效率且影响体系的导电性能,因此,提高传质效率和离子迁移速率,对于提升CSCU工艺的运行效率至关重要。此外,相变过程的稳定运行也依赖于对工况的合理控制,以调节吸收剂的酸度,从而维持相间热力学平衡参数的稳定。

4 结语

硫碳协同资源化CSCU充分发挥原子经济性反应的优势,具有显著的减污增效示范作用。基于非水相铁基离子液低共熔溶液,能够实现H₂S的定向转化和硫磺的高效分离,利用相变吸收技术强化再生组分的富集,促进电解反应过程,提高电化学还原CO₂效率,降低能耗。尽管CSCU面临着诸多挑战,但推动硫碳协同资源化CSCU工艺在能源气开采及化工生产中的实践,对企业的可持续发展,为“双碳”目标的实现具有重要的理论研究和应用意义。

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Summary for “基于离子液低共熔溶液的硫碳协同资源化工工艺的构建与发展”

Synergistic resource recovery of hydrogen sulfide and carbon dioxide in ionic liquid deep eutectic solution: Construction and development

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The efficient capture of hydrogen sulfide (H_2S) and carbon dioxide (CO_2) from acid gases is essential in the chemical industry for achieving ambitious carbon neutrality goals. Transforming H_2S and CO_2 into valuable chemicals is a critical pathway for synergistically reducing pollution and carbon emissions. This transformation not only addresses environmental concerns but also facilitates high-quality progress toward meeting carbon neutrality objectives. The hydrogenation utilization of CO_2 requires a reliable hydrogen supply to react with activated CO_2 molecules. This hydrogen donor can be derived from protons (H) in reaction medium, water (H_2O), and hydrogen atoms, produced by reaction or external supply of H_2 gas.

In the indirect wet oxidation desulfurization process, the H^+ released by H_2S oxidation can be electrolyzed to produce hydrogen. Both hydrogen ions and reactive hydrogen atoms on the electrode surface are crucial precursors for the CO_2 hydrogenation reduction. Using H_2S as hydrogen donor, the functionalized solution system that integrates the indirect wet oxidative desulfurization of H_2S with the electrochemical hydrogenation reduction of CO_2 can be developed to a groundbreaking process known as CO_2 and H_2S capture and utilization (CSCU).

This paper critically reviews CSCU technology in different reaction mediums, including aqueous solutions, organic solvents, and ionic liquids. There are various limitations to aqueous alkaline systems, such as low product selectivity, pronounced hydrogen evolution reactions (HER), and inadequate activity of hydrogen sources. While organic systems can help mitigate some of these issues, their low Faradaic efficiency significantly hampers broader applications. In contrast, iron-based ionic liquid deep eutectic solutions (MIL-DES) present notable advantages in terms of stability, conductivity, and tunability, making them particularly suitable for the advancement of CSCU technical process.

Based on the latest progress in MIL-DES research, this paper proposes a novel CSCU technical process that integrates several critical reactions into one comprehensive process. The innovative process consists of non-aqueous wet oxidative desulfurization, indirect electrolysis of H_2S for hydrogen production, phase change absorption-extraction, and diaphragm-free electrochemical reduction. This multifaceted strategy effectively addresses key problems, including the efficient separation of sulfur during the H_2S oxidation and the improvement of the regeneration process of desulfurization-rich liquid. Furthermore, it enhances the directed overall efficiency of H_2S conversion and CO_2 emission reduction.

However, the development of the CSCU technical process faces multiple challenges. The inherent complexities of raw gas can induce side reactions that make the overall system uncontrolled. Additionally, the desulfurization liquid should possess high thermal and chemical stability to meet the need of long-term operation. Whatever oxidation desulfurization of H_2S or electrochemical reduction of CO , it is necessary to improve the reaction selectivity to match the use of raw gas. Moreover, navigating the intricacies of process control is essential for achieving optimal performance. These challenges require necessitate elevated standards for the development and application of MIL-DES and related technologies.

In short, the successful implementation of the CSCU process promises to deliver dual economic and environmental benefits. This innovative approach not only tackles the pressing challenges of gas capture and conversion but also significantly enhances resource utilization. By paving the way for cleaner energy solutions, the CSCU process holds immense potential and offers extensive application prospects within the energy sector.

sequential H_2S and CO_2 conversion, ionic liquid deep eutectic solution, electrochemical reduction, wet oxidative desulfurization, diaphragm-free electrolysis, phase change extraction

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