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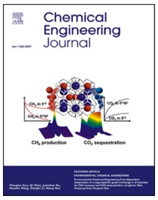

Review

Hydrogen sulfide (H_2S) conversion to hydrogen (H_2) and value-added chemicals: Progress, challenges and outlook

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ABSTRACT

Hydrogen sulfide (H_2S) is a toxic gas released from natural occurrences (such as volcanoes, hot springs, municipal waste decomposition) and human economic activities (such as natural gas treatment and biogas production). Even at very low concentrations, H_2S can cause adverse health impacts and fatality. As such, the containment and proper management of H_2S is of paramount importance. The recovered H_2S can then be transformed into hydrogen (H_2) and various value-added products as a major step towards sustainability and circular economy. In this review, the state-of-the-art technologies for H_2S conversion and utilization are reviewed and discussed. Claus process is an industrially established and matured technology used in converting H_2S to sulfur and sulfuric acid. However, the process is energy intensive and emits CO_2 and SO_2. This calls for more sustainable and energy-efficient H_2S conversion technologies. In particular, recent technologies for H_2S conversion via thermal, biological, plasma (thermal and non-thermal), electrochemical and photocatalytic routes, are critically reviewed with respect to their strengths and limitations. Besides, the potential of diversified value-added products derived from H_2S, such as H_2, syngas, carbon disulfide (CS_2), ammonium sulphate ($(NH_4)_2SO_4$), ammonium thiosulfate ($(NH_4)_2S_2O_3$), methyl mercaptan (CH_3SH) and ethylene (C_2H_4) are elucidated in detail

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Abbreviations: CB, Conduction band; CE, Circular economy; DBD, Dielectric barrier discharge; GAC, Granular activated carbon; GDE, Gas diffusion electrode; GSB, Green sulfur bacteria; HDS, Hydrodesulfurization; IL, Ionic liquid; LCA, Life cycle assessment; MEA, Monoethanolamine; N_2ase, Nitrogenase; NHE, Normal hydrogen electrode; NTP, Non-thermal plasma; PEC, Photoelectrochemical; PVC, Photovoltaic cell; PV-EV, Photovoltaic electrochemical; SAC, Superadiabatic combustion; TEA, Techno-economic analysis; TRL, Technology readiness level; VB, Valence band.

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with respect to the technology readiness level, market demand of products, technical requirements and environmental impacts. Lastly, the technological gaps and way forward for each technology are also outlined.

1. Introduction

In this current era, the development of industries is at an unfathomably rapid state. An excessive amount of essential materials such as fuels, metals and concretes have been consumed to cater towards this development. Fuel is a vital component as the energy precursor. To obtain fuel, crude oil undergoes several processes, producing hydrogen sulfide (H_2S) as a key pollutant. Aside from crude oil processing [1], H_2S also comes from other human economic activities (i.e., natural gas treatment and biogas production) and natural emitters (i.e., hot springs, volcanoes and oceans) [2]. The combined H_2S generation from these sources is estimated to be approximately 7.72 MT per year [3].

H_2S is a fatally hazardous compound due to its toxicity [2,4]. Hence, handling and treating H_2S properly for occupational health and safety preservation is crucial. Exposure limits are set at 10 ppm time-weighted average for application in maritime [5] and construction industries [6]; and 20 ppm (ceiling) for general industry (such as oil and gas industry, petrochemical industry, metal industry, and paper and pulp industry) [7]. Common treatments for H_2S include nitrogen stripping system, using chemical additives such as alkanolamines [8] and aldehyde to remove H_2S from crude oil, and using H_2S scavengers such as glyoxal and quaternary ammonium salt to capture H_2S from crude oil. One of the emerging alternatives to handle H_2S is by converting it to value-added products such as energy and chemicals. To maximize the potential of H_2S conversion, industrial players need to venture into the circular economy (CE) concept. A CE system is specifically developed to integrate a close loop system to effectively recycle by-products or wastes (i.e., H_2S, CO_2, etc.) as a chemical feedstock to produce value-added products (i.e., H_2, O_2, and hydrocarbon) [9–11]. Simultaneously, this allows the development of an effective industrial waste management system, effectively minimizing waste production while generating additional income [10,11].

A thorough understanding of H_2S conversion technologies is required to develop an effective CE system. Studies on H_2S conversion demonstrated an increasing research trend, as indicated in Fig. 1(a). This conveys the relevance and significance of this topic in the current research community. Fig. 1(b) shows that most of these works are published in developed countries such as China, the United States of America, and the Russian Federation, demonstrating the importance and urge to curb H_2S globally. However, review on this subject is extremely limited in the current literature. Thus, this review aims to comprehensively outline all developed and emerging H_2S conversion alternatives for the production of H_2 and other value-added products/chemicals, as well as identify the technical gaps and challenges associated with these conversion techniques, and propose recommendations and insights for future development and commercial deployment.

The review elements are divided into two main sections, (i) Section 2: H_2S conversion to H_2 and value-added products and (ii) Section 3: Challenges and future prospects of H_2S conversion to H_2 and other value-added products. Section 2 discusses all the possible routes of H_2S conversion, including the conversion methods (thermal and biological pathways), thermal and non-thermal plasma, and electrochemical and photocatalytic conversion. Section 3 presents the challenges and outlook of these H_2S conversion routes with respect to their current technology readiness levels (TRLs). Finally, the review ends with a concluding remark in Section 4.

2. H_2S conversion to H_2 and value-added products

2.1. Conventional method

Thermal and biological processes are two conventional methods that have been widely researched for H_2S conversion to value-added products. The most well-known conversion method is the Claus process which utilizes a two-step reaction consisting of thermal oxidation and catalytic reaction to convert H_2S into elemental sulfur [12]. However, the Claus process is associated with several unfavorable drawbacks such as large energy consumption, high capital cost, and high pollutant production [12–14]. This has led researchers to develop various conversion techniques that are more effective and efficient over the years. For example, continuous modification has been made to the existing Claus process, which is discussed in Section 2.1.1. However, newly invented processes such as the superadiabatic combustion (SAC) process by Slimane et al. [15] and the thermal-close loop process by Wang [16] have shown several advantages over the Claus process in terms of production of valuable product such as H_2, lower pollutant emissions and higher energy efficiency [17]. Other than that, biological processes have shown excellent H_2S conversion efficiency of up to 100 % while being environmentally friendly and economically feasible [18,19]. In this section, each process will be elaborated in detail to provide a more in-depth understanding of these processes.

2.1.1. Thermal pathway

Thermal conversion techniques are widely recognized as the most direct alternative with high potential in converting H_2S into value-added

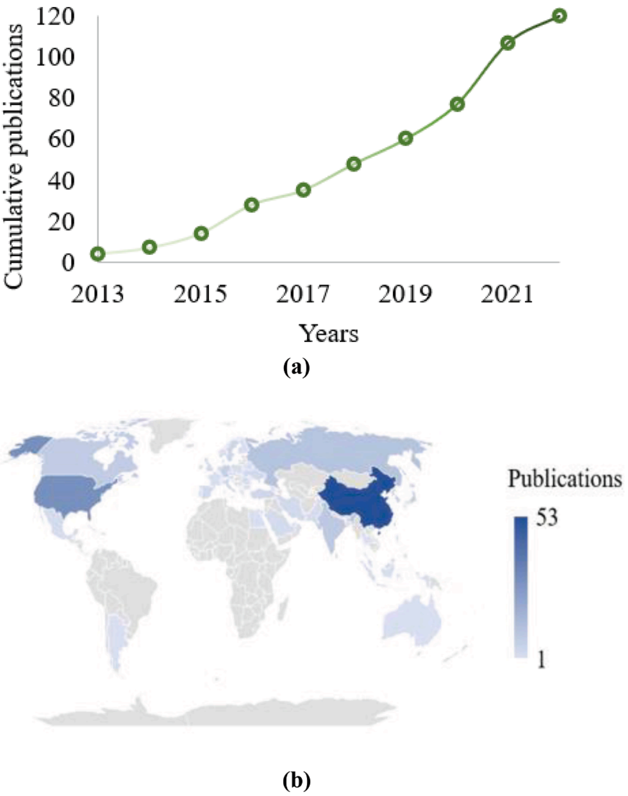

Fig. 1. (a) Cumulative publication of H_2S conversion and utilization research from 2013 to 2022; (b) Publication breakdown based on countries (data extracted from Scopus database).