

Full Length Article

Removal of hydrogen sulfide from coke oven gas by catalytic oxidative absorption in a rotating packed bed



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HIGHLIGHTS

- Catalytic oxidative absorption of H₂S from a simulated COG in a RPB was explored.
- Na₂CO₃ solution doped with a commercial “888” catalyst was used as the absorbent.
- Effect of operating conditions on the removal efficiency of H₂S was studied.
- RPB exhibited higher H₂S removal efficiency than the compared packed column.

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ABSTRACT

The shortage of low-sulfur coking coal will bring about an increasingly high hydrogen sulfide (H₂S) in the coke oven gas (COG) in the near future. This work investigated the removal of hydrogen sulfide (H₂S) from a simulated coke oven gas (COG) by catalytic oxidative absorption in a rotating packed bed (RPB). Sodium carbonate (Na₂CO₃) solution doped with 20 mg L⁻¹ of a commercial “888” catalyst was used as the absorbent. The removal efficiency of H₂S was evaluated under various operating conditions including rotation speed of the RPB (*N*), liquid-gas ratio (*L/G*), inlet H₂S concentration (*c*_{H₂S,in}), temperature (*T*) and Na₂CO₃ concentration (*c*_{Na₂CO₃}) in an attempt to optimize the conditions. The results were validated by comparison with those of separate experiments conducted in a packed column comprising a high-efficiency laboratory packing of Dixon rings. The comparison results reveal that the RPB exhibited higher H₂S removal efficiency than the packed column, indicating it as an efficient gas-liquid contactor with a greater potential to remove H₂S from COG.

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1. Introduction

Coke oven gas (COG), which is a by-product of the coking process, is greatly sought as a high-temperature furnace fuel and city gas by the energy sector in an effort to enhance energy efficiency and minimize emission of greenhouse gas (GHG) [1,2]. In the coking process, a large portion of sulfur in the coal volatilizes into COG, in which 95% of sulfur exists in the form of H₂S [3].

Due to the continued worldwide population and economic demand, the shortage of low-sulfur coking coal compels the producers, especially in developing countries like China which are

short of low sulfur coking coal, to use mixed coking coal of low sulfur and high sulfur or may directly use high-sulfur coking coal in the near future, which consequently results in COG of high H₂S [4–6]. The large presence of H₂S in COG inevitably brings about an array of problems such as catalysts poisoning and equipment corrosion in the industry as well as endangering the environment and human health [1,2,7]. In order to avoid the aforementioned problems associated with the use of COG, the European Standards stipulates that H₂S in COG must be reduced to a level less than 0.5 g m⁻³ [1]. The State Environmental Protection Administration (SEPA) (predecessor of the current Ministry of Environmental Protection) in China has also enforced the Cleaner Production Standard Coking Industry (HJ/T 126-2003), which requires H₂S content in COG must be less than 20 mg m⁻³ for city gas and 200 mg m⁻³ for industrial fuel [8]. More strict regulations are

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Nomenclature

$c_{\text{Na}_2\text{CO}_3}$	concentration of Na_2CO_3 , mol L^{-1}
$c_{\text{H}_2\text{S}, \text{in}}$	inlet H_2S concentration, ppm
$c_{\text{H}_2\text{S}, \text{out}}$	outlet H_2S concentration, ppm
N	rotation speed of RPB, r min^{-1}
$He_{\text{H}_2\text{S}}$	Henry's law constant of H_2S in solution

L/G	liquid gas ratio, L m^{-3}
η	removal of H_2S

expected to be promulgated and enforced by the government in the near future in view of the increasing environment pollution as a result of COG usage. It is therefore necessary to remove H_2S from COG.

The traditional desulphurization technology, which utilizes ammonia for H_2S scrubbing followed by the Clause process, is a well-established technology in the coking industry [9]. However, this approach is associated with some problems. A tail gas treatment equipment is needed to treat about 3–5% of sulfur compound discharged from the Clause process [10]. Moreover, the high content of volatilized NH_3 in acid gas from generation unit will largely consume oxygen to produce water, N_2 , and NO_x in Clause unit, leading to low H_2S removal efficiency and generation of secondary contaminants of NO_x [1,10]. Additionally, about 2% H_2S remaining in ammonia will result into extra SO_2 emission due to the incineration process of ammonia following the regeneration unit [10].

Among other desulfurization technologies, wet oxidation process, which has high desulphurization efficiency and low operation cost, can directly convert H_2S to elemental sulfur with the simultaneous pollution control and resource reclamation [11,12]. This process normally adopts Na_2CO_3 and NaOH as alkali resource, and anthraquinone disulfonic acid (ADA), chelated iron and dinuclear cobalt-phthalocyanine sulfonate (PDS) as catalysts [12,13]. Moreover, the solution can be easily regenerated by air and nearly no air pollutants will be generated. As the development of catalyst with low toxicity, high water solubility and environmental-friendly resource, the potential pollution generated from regeneration and sewage discharge can be efficiently controlled. Commercial “888” catalyst, mainly comprising three nuclear titanium hemicenine cobalt sulfonate ammonium (see Fig. 1), is developed on the basis of PDS catalyst. [12] Compared to PDS catalyst, “888” catalyst is more efficient for the removal of organic sulfur and no co-catalyst is needed. Additionally, this catalyst has an excellent ability to prevent sulfur deposition and avoid the blockage of packing and pipeline. Therefore, “888” catalyst has been used for desulfurization from synthesis gas, COG, fuel gas and biogas [12].

A rotating packed bed (RPB) is a novel multiphase device, which exhibits excellent gas-liquid mass transfer performance, and has been successfully applied to distillation [14], absorption [15,16], wastewater treatment [17,18], VOC Stripping [19] and bromination [20]. Recent studies have successfully employed RPBs in the removal of H_2S by wet approach and RPB with a smaller size compared to traditional packed tower has a promising potential to remove H_2S [21–24]. However, all of these studies mainly focused on treatment of gas containing high H_2S concentration. To the best of our knowledge, the application of RPB in the removal of H_2S from COG containing medium H_2S concentration after the first stage of desulphurization, is still inadequate. This work therefore explored the application of an RPB in removal of H_2S from a simulated COG after the first stage of desulphurization. Sodium carbonate (Na_2CO_3) solution containing 20 mg L^{-1} of a commercial “888” catalyst (mainly comprising three nuclear titanium hemicenine cobalt sulfonate ammonium) was used as the absorbent in the laboratory-scale RPB. The effect of various operating conditions including rotation speed of the RPB (N), liquid-gas ratio (L/G), inlet H_2S concentration ($c_{\text{H}_2\text{S}, \text{in}}$), temperature (T) and Na_2CO_3 concentration ($c_{\text{Na}_2\text{CO}_3}$) on the removal efficiency of H_2S was examined in an attempt to optimize the conditions. Separate experiments were conducted in a packed column comprising a high-efficiency laboratory packing of Dixon rings for a comparison.

2. Reactions for H_2S absorption into Na_2CO_3 solution

The presence of CO_2 (from air) in the simulated COG gas stream resulted into the following reactions [25,26].

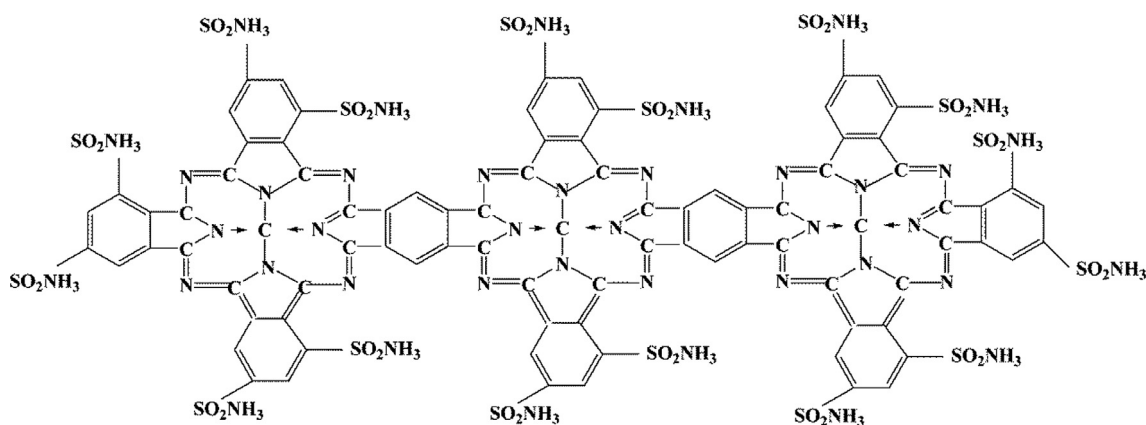
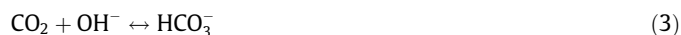


Fig. 1. Molecular structure of three nuclear titanium hemicenine cobalt sulfonate ammonium.

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