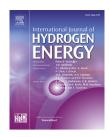
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Evaluation of Bunsen reaction at elevated temperature and high pressure in continuous co-current reactor in iodine-sulfur thermochemical process

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ABSTRACT

Bunsen reaction is one of the three reaction steps of iodine-sulfur process. In present study, Bunsen reaction is carried out in co-current reactor to identify effect of different operating conditions on concentrations of Bunsen reaction product mixture. Bunsen reaction studies have been done in tubular reactor, which is made of tantalum tube and stainless steel jacket, in 50–80 °C temperature range, 2–6 bar (g) pressure range. Feed flow rates are varied for HIx (mixture of hydroiodic acid, water and iodine) 1.2 l/h - 3 l/h, SO₂ 0.02 g/s – 0.24 g/s and O₂ 0.008 g/s –0.016 g/s. It has been observed that, increasing SO₂ feed flow rate and pressure results in increased mole fraction of HI in HIx phase and H₂SO₄ in sulfuric acid phase. Increase in temperature increased the mole fraction of HI in HIx phase but decreased the mole fraction of H₂SO₄ in sulfuric acid phase. Increase in feed I₂/H₂O ratio and HIx feed flow rate, decreased the mole fraction of HI in HIx phase. Higher pressure improved the conversion of Bunsen reactants to products.

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Introduction

Hydrogen is one of the most attractive energy carriers for the future. Hydrogen can play important role to establish clean energy system which is the necessity [1]. If hydrogen is produced from water and not from fossil fuels using nuclear or solar heat, one may achieve a carbon free energy system [2]. Therefore, it is very important to build up the hydrogen production process from water. Large numbers of hydrogen production processes from water are being studied [3]. Thermochemical water decomposition cycle is one of these processes. By combining chemical reactions, water can be decomposed thermochemically at a lower temperature (850 °C) than that required for direct water thermal decomposition (~4000 °C) [4]. The iodine-sulfur (I-S) cycle is one of the most promising thermochemical water splitting technologies for the massive production of hydrogen [5]. This cycle was originally developed in the 1980s by General Atomics (GA) and relied on the high temperature thermal energy of nuclear reactors [6].

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I-S cycle consists of three sections as expressed in Eqs. (1)-(3).

$$2H_2O + I_2 + SO_2 \iff H_2SO_4 + 2HI$$
 (1)

 $H_2SO_4 \longrightarrow H_2O + SO_2 + \frac{1}{2}O_2$ (2)

2HI
$$\longrightarrow$$
 I₂ + H₂ (3)

The first process expressed by Eq. (1) is the Bunsen section, where the water reacts with sulfurdioxide and iodine at

Table 1 – Merits and demerits of different modes of Bunsen reactor operation.		
Mode of operation	Merits	Demerits
Co current	Presence of excess I_2 throughout the reactor thus avoids the side reactions Easier to scale up Heat removal is easy at any scale	Leads to more number of equipments in the I-S process flow sheet
Counter current	Leads to less number of equipments in the flow sheet as reaction and phase separation are integrated in the same equipment	Scale up is difficult
	Multi stage contact	Presence of flooding at high liquid throughputs Issue with respect to heat removal at large scale

relatively low temperatures, and the product mixture is spontaneously separated into the sulfuric acid (SA) and HIx phases (mixture of HI, H₂O, I₂) in the presence of excess iodine [6,7]. The SA phase is fed to the second process, the SA decomposition section, where the SA is concentrated by distillation and then decomposed using catalyst into oxygen and sulfurdioxide by heating in the highest temperature range of the I-S cycle [8], as mentioned in Eq. (2). The sulfurdioxide and water are recycled to the Bunsen section after removing heat. The HIx phase is sent to the third process, the hydrogen iodide (HI) decomposition section, where the HI is separated from the mixture by distillation, and decomposed using catalyst into hydrogen and iodine by heating at intermediate temperatures [9], as shown in Eq. (3). Membranes are also used for HI decomposition [10,11].

Increase in Bunsen reaction operating temperature leads to decrease in SO₂ solubility [12]. Therefore to increase the SO₂ solubility in Bunsen reaction solution, higher pressures are required. Higher pressure operating conditions are favourable for the integrated operation of the Bunsen reaction. Increase in pressure shifts the HI azeotrope to higher value and reduces the pumping power required to compress hydrogen which is produced [13]. Bunsen reaction and phase separation has been studied extensively under various operating conditions [12,14-29]. Kim HS et al. carried out Bunsen reaction by varying temperature, pressure, feed iodine and feed water concentrations in semi batch reactor, extent of Bunsen reaction was improved by increasing pressure and initial H₂O/HI concentration [14]. Rao AS et al. conducted series of experiments in semi batch reactor to study Bunsen reaction and concluded that the overall reaction rate was increased with pressure and decreased with temperature [19]. Bunsen

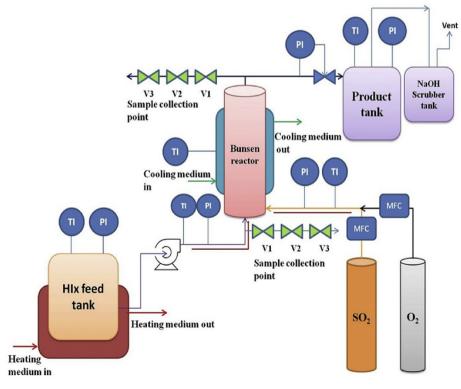


Fig. 1 – Schematic of Bunsen reaction experimental setup.

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