Regeneration behavior of tin oxide sorbent for warm syngas desulfurization

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A R T I C L E   I N F O

Article history:
Received 31 May 2017
Revised 31 July 2017
Accepted 31 August 2017
Available online 23 September 2017

Keywords:
SnO₂
Regeneration
Steam
SO₂
Tin sulfide
Sintering

A B S T R A C T

The steam regeneration and SO₂ regeneration of tin oxide for warm syngas desulfurization is studied in the temperature range of 400–600 °C. In the steam regeneration, reversible removal of H₂S achieved. Regenerated H₂S concentration increased with the increasing regeneration temperature. SnO₂ sorbent can achieve a complete regeneration by steam at 500 and 600 °C. In the SO₂ regeneration, elemental sulfur was produced by the reaction of SnS and SO₂. Raising the regeneration temperature (500–600 °C) or SO₂ concentration (1.5–10 vol%) improved the regeneration rate. Under SO₂ regeneration at 500 °C, SnS₂ formed in the sorbent due to the interconversion of tin sulfides. Under steam regeneration or SO₂ regeneration, the cyclic breakthrough sulfur capacity of SnO₂ sorbent decreased because of the sintering caused by low melting component SnS. A two-stage regeneration process was applied to recover the elemental sulfur which achieved a complete regeneration.

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1. Introduction

Hydrogen production from coal syngas based on H₂/C₀₂ separation after the water gas shift reaction is a promising technology for advanced coal utilization due to its advantages of high efficiency and low emissions. However, coal syngas usually contains 0.2%–1.0% H₂S and other sulfur compounds [1]. Desulfurization is a crucial process because H₂S is not only harmful to human health but also causes catalyst poisoning and equipment corrosion. Hot syngas desulfurization uses metal oxides or mixed metal oxides as sorbent and the process consists of sulfidation and regeneration. The regeneration process is critical for the sorbent selection. There are three ways to regenerate the sulfided sorbents with different sulfur products.

Oxygen regeneration:

\[ \text{Mes}_m + (m + 0.5x)O_2 = \text{MeO}_x + m \text{SO}_2 \]

Steam regeneration:

\[ \text{Mes}_m + yH_2O = \text{MeO}_y + mH_2S + (y - m)H_2 \]

SO₂ regeneration:

\[ \text{Mes}_m + 0.5z\text{SO}_2 = \text{MeO}_z + (m + 0.5z)S \]

Oxygen is the most common way for the regeneration. ZnO [3], CuO [4] and MnO [5] can be regenerated under O₂ atmosphere. However, the reaction is very exothermic. In order to control the regeneration temperature, a diluted oxygen or air must be used which results in the diluted SO₂ [2]. SO₂ is another pollutant and requires further treatment. The formation of sulfate is also unfriendly to oxygen regeneration. H₂S is the product in the steam regeneration. Walcker et al. chose Mn/γ-Al₂O₃ and Fe/γ-Al₂O₃ as candidates for steam regeneration [6]. For Mn/γ-Al₂O₃, MnAl₂O₄ formed during the preparation. Compared with MnO, MnAl₂O₄ absorbed H₂S more weakly which could be regenerated with H₂O. However, the sensitivity of H₂O in the sulfidation and large amounts of steam consumption in the regeneration are two major drawbacks. The direct production of elemental sulfur is possible in the SO₂ regeneration. Bakker et al. tested Mn-based sorbent regeneration by SO₂ to recover elemental sulfur, but the regeneration was not complete [7]. An excess diluted O₂ regeneration was needed to regenerate the strong sites. The research of Zeng et al. pointed out that reduced CeO₂ was efficient for hot syngas desulfurization [8]. The sulfided product, Ce₂O₅ reacted with SO₂ to produce elemental sulfur directly over the range of 500–700 °C. Compared with Mn-based sorbents, SO₂ regeneration of Ce-based sorbent was rapid and complete. Fe₂O₃ has a poorer desulfurization performance than ZnO, but it is superior in the regeneration process. All the mentioned regeneration methods could
be applied to regenerate the sulfided Fe-based sorbents [10–12]. In the steam regeneration, the main product was Fe$_2$O$_4$ and H$_2$S, while in the SO$_2$ regeneration, elemental sulfur was obtained and Fe$_2$O$_4$ was slowly oxidized to Fe$_2$O$_3$. The direct elemental sulfur production by SO$_2$ regeneration is more attractive which avoids the further treatment of sulfur product.

In the previous studies, SnO$_2$ is not within the scope of research because SnO$_2$ is reduced to a low-melting tin in a high reducing atmosphere at the high temperature [13]. Droog et al. suggested that desulfurization operated at moderate temperatures of 343–538 °C could be better because the capital and operation cost was lower and the overall IGCC process efficiency improved only a little [14]. At moderate temperatures, the reduction of SnO$_2$ will not be a big problem. SnO$_2$ has a favorable regeneration characteristic that both steam regeneration and SO$_2$ regeneration are possible for SnO$_2$ sorbent [15,16]. Haldor Topsoe (Denmark) used a high pressure steam to regenerate SnS at 500 °C and received a 2–3% H$_2$S in the outlet. They did not focus on the detailed study of steam regeneration characteristics. TDA (USA) took a SO$_2$ containing gas from the regeneration of ZnS by diluted O$_2$ to the SnS bed and elemental sulfur was obtained at 700 °C. However, SnO$_2$ occupied the lower part of the sorbent bed in their study, so the concentration of regeneration gas for SnO$_2$ contained excess O$_2$ and SO$_2$ was uncertain.

The objective of this work was to study the regeneration performance of SnO$_2$ sorbent and present the characteristics of steam regeneration and SO$_2$ regeneration. The influence of regeneration operation factors (i.e. temperature) and the physical and chemical properties of SnO$_2$ sorbent after regeneration were investigated. The cyclic desulfurization performance of SnO$_2$ sorbent and the improvement of the regeneration method were also discussed.

2. Experimental

2.1. Preparation of sorbents

SnO$_2$ sorbent used in the experiments was an analytical reagent from Sinopharm Chemical Reagent. Before sulfidation tests, SnO$_2$ sorbent was pressed and sieved through 97–150 μm. The commercial SnO$_2$ was treated in a 100 mL/min 50 vol% H$_2$, and balanced N$_2$ flow at 400–500 °C for 3 h. The treated sorbent was analyzed by X-ray photoelectron spectroscopy (XPS).

Table 1. Regeneration conditions used in regeneration tests.

<table>
<thead>
<tr>
<th>Steam regeneration</th>
<th>SO$_2$ regeneration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>400, 500, 600</td>
</tr>
<tr>
<td>Pressure (MPa)</td>
<td>0.1</td>
</tr>
<tr>
<td>Flowrate (mL/min)</td>
<td>60</td>
</tr>
<tr>
<td>Gas composition</td>
<td>H$_2$O: 50 vol% N$_2$: balance</td>
</tr>
</tbody>
</table>

2.2. Characterization of sorbents

The XPS analyses were performed using an ESCALAB 250Xi spectrometer manufactured by Thermo Fisher. A non-monochromatic Al Kα X-ray source was used, and samples were analyzed as pellets. The phase composition analysis of the sorbents using X-ray diffraction (XRD) was determined on a Rigaku D-max 2500 X-ray diffractometer with Cu Kα radiation. The experimental data was digitally collected by a “step by step” scanning method in a 2θ angle interval of 10–70°. The textural properties of the sorbents using Brunauer–Emmett–Teller (BET) theory and Barret–Joyner–Halenda (BJH) model were performed on an ASAP 2020 (Micromeritics, USA) by determining the nitrogen adsorption-desorption isotherms at −196 °C. The sorbent surface morphology was determined by a scanning electron microscopy (ZEISS MERLIN VP compact, Germany).

2.3. Sulfidation and regeneration tests of SnO$_2$

The sulfidation and regeneration tests of SnO$_2$ were evaluated in a fixed-bed reactor with an inside diameter of 10 mm. 1.0 g of SnO$_2$ was charged into the reactor and heated to 400 °C in a N$_2$ atmosphere. Before the sulfidation, SnO$_2$ was reduced by a 50 vol% H$_2$ and balanced N$_2$ for 1 h. Then, a feed gas containing 0.5 vol% H$_2$S, 50 vol% H$_2$ and balanced N$_2$ was introduced from the top of the reactor with a flowrate of 60 mL/min. The concentrations of H$_2$S in the inlet and outlet were analyzed by a gas chromatograph (GC-9A, Shimadzu) equipped with a flame photometry detector (FPD). Since GC-9A was applied for low concentration analysis, an inert N$_2$ was used to dilute the gas concentration to 1/10 of the original gas concentration. The breakthrough point (BT point) was defined as the time when the outlet H$_2$S concentration reached 50 ppmv, which is 1% of the inlet H$_2$S concentration in the feed gas. The amount of sulfur absorbed by the sorbent was calculated by the following equation:

$$q = \frac{32}{22.4} \times \frac{Q_{\text{sulfidation}}}{M_{\text{sorbent}}} \times \int_0^{t_1} (x_{\text{in}} - x_{\text{out}}) \, dt$$

After sulfidation, the sulfided sorbent was regenerated at 400–600 °C. The regeneration conditions are presented in Table 1. In the steam regeneration, the steam was produced by heating the water and the flowrate of water was controlled by a pump (LabAlliance). The outlet gas flew through the screwed quartz tube in the ice water to remove steam and analyzed by GC-9A. The H$_2$S recovery $R_{\text{H}_2\text{S}}$ was obtained by the following equation:

$$R_{\text{H}_2\text{S}} = \frac{\frac{32}{22.4} \times 50\% \times Q_{\text{regeneration}} \times \int_0^{t_2} x_{\text{re,dry}} \, dt_2}{M_{\text{sorbent}} \times q} \times 100\%$$

$$= \frac{50\% \times Q_{\text{regeneration}} \times \int_0^{t_2} x_{\text{re,dry}} \, dt_2}{Q_{\text{sulfidation}} \times \int_0^{t_1} (x_{\text{in}} - x_{\text{out}}) \, dt_1} \times 100\%$$

In SO$_2$ regeneration, the outlet gas flew through the U-tube with quartz wool in the ice water to condense the elemental sulfur and analyzed by GC-9A. The condensed sulfur in the U-tube
was collected with quartz wool and weighed. The gained weight was the weight of produced elemental sulfur. The elemental sulfur recovery \( R_s \) was calculated based on the following reaction:

\[ \text{SnS} + \text{SO}_2 = \text{SnO}_2 + \text{S}_2 \]

So the elemental sulfur recovery \( R_s \) can be obtained:

\[ R_s = \frac{m_s/2}{M_{\text{sorbent}} \times q} \times 100\% \]

During the cyclic desulfurization tests, 2 g of the sorbent was charged and the regeneration process was finished until the \( R_{H_2S} \) and \( R_s \) reached 100% except for the SO2 regeneration at 500 °C. When the regeneration test was finished, the reactor was cooled to 400 °C and purged with N2 for the next sulfidation.

3. Results and discussion

3.1. Steam regeneration

3.1.1. Sulfidation of SnO2 sorbent at 400 °C

Fig. 1 shows Sn 3d XPS spectra of SnO2 under different temperature treatment. The binding energy values of Sn 3d at 484.0 eV and 492.5 eV were assigned to Sn0 [17]. It meant that after 50 vol% H2 treatment at 400 °C, there was no metallic Sn in SnO2 sorbent. The commercial SnO2 sorbent can be used for the sulfidation at 400 °C.

Before the regeneration test, the sulfided sorbents were prepared by the reaction with 0.5 vol% H2S in a reducing atmosphere at 400 °C. Fig. 2 shows the typical sulfidation breakthrough curve of SnO2 sorbent at 400 °C. Before the breakthrough, the sorbent removed nearly 100% of the H2S with less than 1 ppmv H2S detected by GC. After breakthrough, the rate of sulfidation went slow because the slope of outlet H2S concentration curve became flat. The XRD results in Fig. 3(a) and (b) showed that the main species in the reduced sorbent was SnO2 (PDF: 41-1445) and the sulfided sorbent exhibited typical peaks which were assigned to SnS (PDF: 39-0354). Besides, the peaks of SnO2 were also observed. The total absorbed sulfur amount in 420 min was 157.4 mg, which was about 74% of the theoretical sulfur capacity of SnO2. At the conclusion of the sulfidation process, majority of SnO2 turned into SnS which could be used for the regeneration test.

3.1.2. Effect of temperature on steam regeneration

The outlet H2S concentration curves of SnO2 sorbent at different temperatures are shown in Fig. 4. H2S was the sulfur product obtained in the steam regeneration. When the regeneration temperature was 400 °C, about 1100 ppmv H2S was detected in the outlet gas which was stable in 1200 min with a tiny decrease. The
regeneration was very slow at 400 °C, and the sorbent could not be regenerated completely in 1200 min. When the regeneration temperature increased to 500 and 600 °C, the time for complete regeneration decreased, as shown in Table 2. The H₂S concentration was higher with the higher regeneration temperature. At 600 °C, the H₂S concentration was 6500 ppmv in the initial period. The decrease of the H₂S concentration also became quicker which led to a short regeneration time. Raising the regeneration temperature increased the reaction rate of SnS and H₂O and reduced the amount of steam consumption. However, the high temperature steam was expensive and not easy to obtain. Fig. 5 shows the XRD patterns of sorbents regenerated by steam at 500 and 600 °C. After steam regeneration, no sulfided phase (ca. SnS) was detected and the crystal structure turned back to cassiterite SnO₂ again. The calculated H₂S recoveries were 93.97% and 85.65% at 500 and 600 °C, respectively. A small amount of H₂S might be dissolve in the water because the pH value of the condensed water decreased. Steam regeneration achieved a complete conversion of SnS back to SnO₂ at 500 and 600 °C.

In the steam regeneration, high concentration H₂S is what we are concerned. As nitrogen is an inert component in the regeneration gas, the concentration of nitrogen can be reduced to increase the H₂S concentration in the regeneration gas. For further utilization, the high concentration H₂S can be converted into elemental sulfur by Claus process. However, as shown in Table 2, the steam regeneration time is very long with the large amount of steam consumption in the regeneration.

3.2. SO₂ regeneration

3.2.1. Effect of temperature on SO₂ regeneration

The SO₂ regeneration curves of SnO₂ sorbent at different temperatures are shown in Fig. 6. When SO₂ was taken as a regeneration gas, elemental sulfur condensed at the outlet of the reactor. t* was 120 min. In order to evaluate the production rate of elemental sulfur in the regeneration, the condensed sulfur at the outlet of the reactor was collected at every 120 min. At 500 °C, the sulfur recovery increased slowly to about 40% in 720 min. The SO₂ regeneration rate was relatively slow at 500 °C and most of the inlet SO₂ was not used in the regeneration process. Because elemental sulfur condensed in the outlet, SO₂ could be recycled to regenerate the sorbent. When the temperature increased to 550 and 600 °C, the sulfur recovery increased notably. With the increasing reaction rate, the regeneration time decreased. At 600 °C, the sulfur recovery reached 70.87% in t*.

3.2.2. Effect of SO₂ concentration on SO₂ regeneration

The SO₂ regeneration curves of SnO₂ sorbent at different SO₂ concentrations are shown in Fig. 7. At 500 °C, the regeneration rate was very slow at 1.5 vol% SO₂. When SO₂ concentration increased to 5 vol%, elemental sulfur recovery increased from 34.43% to 58.45% in the 10 h. If SO₂ concentration was further increased to 10 vol%, the improvement of elemental sulfur recovery was not obvious with 63.64% in 600 min. Raising SO₂ concentration in the regeneration gas was beneficial to the elemental sulfur recovery. However, there was a limitation in the improvement by the SO₂ concentration. Compared with 1.5 vol% SO₂ at 600 °C, the regeneration rate was still slow when 10 vol% SO₂ used as the regeneration gas at 500 °C.

3.2.3. Chemical properties of SnO₂ sorbent after SO₂ regeneration

Fig. 8 shows the XRD patterns of the sorbents after 1.5 vol% SO₂ regeneration. After SO₂ regeneration at 600 °C, SnO₂ was the only crystal phase in the regenerated sorbent. Tin sulfide phase was no longer detected by XRD. Regeneration at 600 °C can reach a deep regeneration of the sorbent which was coincided with the elemental sulfur recovery. After SO₂ regeneration at 500 °C, SnO₂ was the main crystal phase in the sorbent. Besides, there were some peaks assigned to SnS₂ [PDF: 22-0951]. After sulfuration in the reducing atmosphere, SnS was the only crystal phase of tin sulfide in the sorbent, as shown in Fig. 3(b), indicating that after SO₂ regeneration at 500 °C, a part of SnS converted into SnS₂. There was another interesting phenomenon in the SO₂ regeneration. Table 3 lists the elemental sulfur recovery in 0–120 min and 120–240 min under different SO₂ regeneration conditions. Elemental sulfur recovery in 0–120 min was lower than that in 120–240 min under low SO₂ concentration and low regeneration temperature. This unusual phenomenon could be explained with the formation of SnS₃ in the SO₂ regeneration. There are several forms of tin sulfides, ca. SnS, Sn₂S₃, Sn₂S₅. They could interconvert into each other at different temperatures [18]. At lower temperatures, ca. 500 °C, the following reaction occurred:

SnS + S = SnS₂

The regeneration test was conducted in a fixed-bed reactor. In the upper zone, the formation of elemental sulfur happened under SO₂ gas flow. This newly formed gaseous sulfur moved downstream, and reacted with SnS to form SnS₂. Due to the consumption of elemental sulfur in the lower zone, very limited sulfur could move to the outlet in the beginning. As the reaction carried out, the amount of elemental sulfur increased in the outlet. With the increase of SO₂ concentration and regeneration temperature, the formation rate of sulfur increased and the formation rate of SnS₂ decreased at high temperatures, also the time of this unusual phenomenon decreased. So we did not observe it in the comparison of 0–120 min and 120–240 min. Sulfate formation is another important problem in many cases of SO₂ regeneration. Because SnSO₄ decomposes into SnO₂ and SO₂ when the temperature is higher than 500 °C [19]. No SnSO₄ would
be formed in the regenerated sorbent which is confirmed by XRD results.

3.3. Cyclic desulfurization performance of SnO₂ sorbent

3.3.1. Under steam regeneration

As shown in Fig. 9, after steam regeneration at 500 °C, the desulfurization performance of the sorbent decreased. The BT sulfur capacity dropped from 141.4 mg/g to 36.4 mg/g in 3 cycles. From 3rd cycle, the decrease of the BT sulfur capacity became slow. In these cycles, after the initial H₂S breakthrough, H₂S concentration in the outlet began to increase in a relatively slow rate, which meant that the sorbent still reacted with H₂S in a slow reaction rate. During the steam regeneration, the sorbent became unfavor-

able to H₂S. Steam regeneration at 500 °C achieved a complete conversion of SnS back to SnO₂. Therefore, the physical properties of the sorbent before and after desulfurization were characterized and the results are shown in Table 4 and Fig. 10.

The BET specific surface area (SSA) of fresh SnO₂ sorbent was 6.01 m²/g. During the sulfidation process, pores in the sorbent

### Table 3. Comparison of elemental sulfur recovery (Rₚ) of SnO₂ sorbent at different SO₂ regeneration conditions.

<table>
<thead>
<tr>
<th>Regeneration conditions</th>
<th>500 °C, 1.5 vol% SO₂</th>
<th>550 °C, 1.5 vol% SO₂</th>
<th>600 °C, 1.5 vol% SO₂</th>
<th>500 °C, 5 vol% SO₂</th>
<th>500 °C, 10 vol% SO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rₚ [%] 0–120 min</td>
<td>2.97</td>
<td>17.63</td>
<td>70.87</td>
<td>4.43</td>
<td>22.15</td>
</tr>
<tr>
<td>120–240 min</td>
<td>8.80</td>
<td>26.42</td>
<td>19.00</td>
<td>17.41</td>
<td>21.52</td>
</tr>
</tbody>
</table>

### Table 4. BET SSA and pore volume of fresh SnO₂, 1st and 6th regenerated sorbents.

<table>
<thead>
<tr>
<th>SnO₂ samples</th>
<th>Fresh</th>
<th>1st regenerated</th>
<th>6th regenerated</th>
</tr>
</thead>
<tbody>
<tr>
<td>BET SSA (m²/g)</td>
<td>6.01</td>
<td>2.70</td>
<td>2.19</td>
</tr>
<tr>
<td>Pore volume (mL/g)</td>
<td>0.034</td>
<td>0.011</td>
<td>0.007</td>
</tr>
</tbody>
</table>
Fig. 8. XRD patterns of SnO$_2$ sorbents after 1.5 vol% SO$_2$ regeneration.

Fig. 9. Cyclic breakthrough curves for 2 g SnO$_2$ sorbent by steam regeneration at 500 °C.

Fig. 10. SEM images of fresh (a) and 6th regenerated (b) SnO$_2$ sorbent.
were blocked. After steam regeneration, the BET SSA decreased to 2.19 m²/g, and pore volume was 0.007 mL/g. It indicated that the damage of the pore structure in the SnO₂ sorbent was irreversible during the cyclic tests. On one hand, the densities of SnO₂ and SnS were 6.95 and 5.22 mL/g, respectively. Thus, the sulfide product SnS occupied more space than the sorbent. Within the cyclic sulfidation and regeneration, the repeated expansion and contraction would cause the increasing pore diameter in the sorbent [20]. Fig. 10(a) and (b) shows the aggregation of SnO₂ particles and the pores became larger. On the other hand, the melting point of SnS is 882 °C. When the temperature was above Tammann temperature, sintering happened in the sorbent [21]. Since the melting temperature of SnS was low (Tammann temperature is approximately 0.5 of the melting point in absolute units), the sintering happened in the sulfided sorbent during the reaction temperature of 400–600 °C. Sintering caused the loss of pore which was confirmed by many sorbents [22]. Due to the sintering of SnO₂ sorbent in the desulfurization process, the BET SSA decreased and pore structure damaged with low pore volume. The degradation of the pore structure resulted in quick decrease of BT sulfur capacity in the first 3 cycles.

### 3.3.2. Under SO₂ regeneration

Fig. 11 shows the cyclic breakthrough curves for 2 g SnO₂ sorbent under 1.5 vol% SO₂ regeneration. For SO₂ regeneration at 600 °C, after a complete regeneration with nearly 100% elemental sulfur recovery, the cyclic BT sulfur capacity dropped from 135.0 to 51.4 mg/g in three cycles. The decrease of BT sulfur capacity existed in both regeneration methods. The SO₂ regeneration at 500 °C was not complete with a 42.91% of elemental sulfur recovery. In the SO₂ regeneration, two shortages should be concerned. First, the formation of SnS₂ appeared at low regeneration temperatures, i.e. 500 °C. Second, it was difficult to determine the terminal of the SO₂ regeneration because the sulfur product was solid sulfur and the change in the SO₂ concentration would be small at the end of the regeneration. Fig. 12 shows the breakthrough curves of incomplete regenerated SnO₂. A high concentration H₂S was detected in the outlet during the reduction period and H₂S removal efficiency dropped. Within the formation of SnS₂, due to the reaction between H₂ and SnS₂ [22], the amount of produced H₂S increased. It indicated that the sorbent through incomplete SO₂ regeneration was not available for cyclic removal of H₂S. In order to avoid this drawback, a new regeneration method with two stages was applied to SnO₂ sorbent regeneration, as shown in Fig. 13.

The sorbent was mainly regenerated by SO₂ and elemental sulfur was recovered. The sulfur recovery depended on the regeneration temperature and SO₂ concentration. Then, a steam purged through the sorbent bed to make sure that all the sorbents were completely regenerated. As shown in Fig. 12, after the steam regeneration at 500 °C, the sorbent kept the ability for H₂S removal. There were several advantages of this two-stage regeneration. First, elemental sulfur was a more desirable product for the desulfurization process and the steam consumption was significantly reduced. Second, because the complete regeneration of SnO₂ sorbent was very important for cyclic utilization, H₂S concentration in the steam regeneration can be the signal for determining the complete regeneration of the sorbent. Third, the two-stage regeneration could adjust the ratio of elemental sulfur to H₂S according to the actual demands.
4. Conclusions

The steam regeneration and SO\(_2\) regeneration of SnO\(_2\) sorbent were studied at 400–600 °C. SnO\(_2\) was not reduced to metallic tin but sulfided into SnS at 400 °C. H\(_2\)S and elemental sulfur were obtained in the steam and SO\(_2\) regeneration, respectively. Raising the regeneration temperature was beneficial to both the steam and SO\(_2\) regeneration. Increasing SO\(_2\) concentration also improved the recovery of elemental sulfur.

Steam regeneration achieved a complete regeneration for SnO\(_2\) sorbent with the long regeneration time and a large amount of water consumption. SnS\(_2\) appeared in the SO\(_2\) regeneration at low temperature (i.e., 500 °C) due to the interconversion of tin sulfides. The cyclic breakthrough sulfur capacity of SnO\(_2\) sorbent decreased from 141.4 to 23.6 mg/g in the first three cycles. The low melting component SnS caused sintering in the reaction temperature of 400–600 °C. A new regeneration process combining with SO\(_2\) and steam regeneration were designed and analyzed to recover elemental sulfur and achieve a complete regeneration.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (51476092), the National High Technology Research and Development Program of China (2011AA060501) and Shanxi Province Science and Technology Major Programs (MH2015-06).

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