

A Study on Carbothermal Reduction of Sulfur Dioxide to Elemental Sulfur Using Oilsands Fluid Coke

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Experiments and reaction equilibrium calculations were carried out for the SO₂ gas and oilsands fluid coke system. The goal was to develop a coke-based sulfur-producing flue gas desulfurization (SP-FGD) process that removes SO₂ from flue gases and converts it into elemental sulfur. The conversion of SO₂ to elemental sulfur proceeded efficiently at temperatures higher than 600 °C, and the sulfur yield reached a maximum (>95%) at about 700 °C. An increase of temperature beyond 700 °C enhanced the reduction of product elemental sulfur, resulting in the formation of reduced sulfur species (COS and CS₂), which lowered the sulfur yield at 900 °C to 90%. Although equilibrium calculations suggest that a lower temperature favors the conversion of SO₂ as well as the yield of elemental sulfur, experiments showed no formation of elemental sulfur at 600 °C and below, likely due to hindered kinetics. Faster reduction of SO₂ was observed at a higher temperature in the range of 700–1000 °C. A complete conversion of SO₂ was achieved in about 8 s at 700 °C. Prolonging the product gas–coke contact, the yield of elemental sulfur decreased due to the formation of COS and CS₂ while the SO₂ conversion remained complete. Equilibrium calculations suggest that the ultimate yield of elemental sulfur maximizes at the C/SO₂ ratio of 1, which represents the stoichiometry of SO₂ + C → CO₂ + S. For the C/SO₂ ratio < 1, equilibrium calculations predict elemental sulfur and CO₂ being major products, suggesting that SO₂ + C → CO₂ + S is the predominant reaction if SO₂ is in excess. Experiments revealed that elemental sulfur and CO₂ were only major products if the conversion of SO₂ was incomplete, which is in agreement with the result of the equilibrium modeling.

Introduction

Sulfur dioxide, the most important precursor of acid rain, is being produced in vast quantities by fossil fuel-powered utility plants and nonferrous metal smelters worldwide. Although significant progress has been made in SO₂ emission control in the last few decades, acid rain is still an environmental concern in many parts of the world, even in developed countries such as Canada and the United States (1, 2). Since SO₂ abatement is an expensive practice, the key challenge

is to develop low-cost technologies. Commercial flue gas desulfurization (FGD) processes are mostly of the throwaway type in which alkaline materials react with SO₂ to form alkaline metal sulfate that is often stockpiled (3). To minimize the waste disposal problem associated with the existing technologies, a number of alternatives have been suggested for the recovery of sulfur from SO₂. The common forms of the recovered sulfur are sulfuric acid, liquid sulfur dioxide, and elemental sulfur. Sulfuric acid is perhaps the most important inorganic chemical in terms of the amount produced and consumed by a wide range of industries, while liquid sulfur dioxide is used in relatively small quantities by certain industries such as pulp and paper. Elemental sulfur can be utilized as a raw material for essentially all sulfur-containing products and is sometimes desirable since it is easier to store and transport (4, 5).

The key part in sulfur-producing flue gas desulfurization (SP-FGD) technologies is the reduction of sulfur dioxide to elemental sulfur. Hydrogen and carbon monoxide have been successfully used as reducing agents for SO₂ in the presence of catalysts. However, no H₂- or CO-based technology has been commercialized (6) due to the high cost involved in the production and transportation of these gases. Carbon in various forms, such as coal and charcoal, was examined as a reducing agent for SO₂ extensively (4). Biswas et al. (7) reduced SO₂ to carbon disulfide (CS₂) and elemental sulfur with various types of coal and concluded that the volatile hydrocarbons present in these coals reduced the yield of elemental sulfur due to the formation of H₂S. Blackwood and McCarthy (8) examined the reaction of chars from brown coal with SO₂ and obtained high selectivity to either elemental sulfur or CS₂ by varying conditions. Steiner et al. (9) reported a process with anthracite as the desired reductant, and based on this study, the Foster Wheeler Co. developed the RESOX process (10). Ratcliffe and Pap (11) reported high yields (90%) of elemental sulfur with low-rank coal and lignite. In most of these studies, minimizing volatile matters was desirable as it suppressed the formation of H₂S and other undesirable products. In their patent, Steiner et al. (12) indicated that both SO₂ conversion and elemental sulfur yield increased as the content of the recycled coal was increased. The recycled coal was the coal that had been exposed to high temperature and therefore had less volatile matter. This suggests that oilsands fluid coke could be a more suitable carbon source than coal. It should be pointed out that coal is usually more reactive at low temperatures than coke due to higher volatile hydrocarbon contents. Recently, a wet SP-FGD process was developed based on simultaneous SO₂ absorption and H₂S generation using aqueous sulfide (13). In this process, elemental sulfur is produced via Claus reaction, and carbon is used for the regeneration of sulfide from sulfite (14).

Oilsands fluid coke is a byproduct in the conversion of bitumen to synthetic crude oil and is being produced and stockpiled in large quantities. Chung et al. (15) reported that about 5000 ton/day of the coke is being stockpiled in the proximity of the Syncrude plant in northern Alberta, Canada. Containing over 85 wt % carbon, the oilsands fluid coke represents a significant energy resource. Its utilization as a solid fuel is limited by its high sulfur content (6–8 wt %), which is however not a concern if used as a reducing agent in an SP-FGD process. The purpose of this study is to investigate experimentally the feasibility of using oilsands fluid coke for carbothermal reduction of SO₂ to elemental sulfur. Equilibrium calculations were carried out to determine theoretical limits of the C–SO₂ system and to assist the experimental design of operating conditions. It is also hoped

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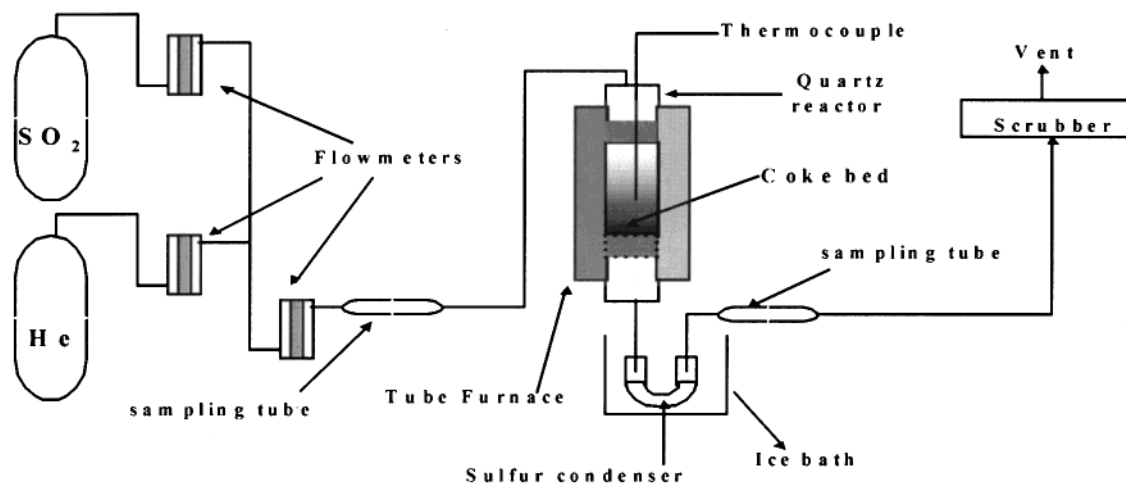


FIGURE 1. Experimental setup.

that by combining experiments with the equilibrium reaction calculations, the controlling mechanism of carbothermal reduction of SO_2 with oilsands fluid coke will be better understood.

Experimental Section

Figure 1 shows the experimental setup consisting of three separate sections: gas supply, main reactor, and product sampling. High-purity SO_2 and helium were supplied from compressed gas cylinders to flowmeters before entering a gas mixer. The flow rate of the gas mixture was measured, and the concentration was monitored using a gas chromatograph. SO_2 concentration was kept constant at 15 vol % for all runs. The reactor was a 14 mm i.d., 1.5 mm thick, and 600 mm long quartz tube placed vertically in a tubular furnace with an automatic temperature controller. The reactor consisted of two zones. The reaction zone was packed with fluid coke particles (usually in grams). The discharge or outlet zone was packed with quartz chips to reduce the residence time of gaseous products in the discharge zone and the reactor and therefore minimize secondary reactions among products. The fluid coke was charged from the top of the reactor and supported by a porous quartz disk placed 150 mm above the bottom of the reactor. The gas mixture traveled downward through the packed coke bed. The coke bed was heated to a set temperature (400–1000 °C) with purged helium before the SO_2 was introduced at the start of the runs. The reaction temperature was monitored with a thermocouple in the middle of the coke bed. The exit gas, from the bottom of reactor, passed through a sulfur collector/condenser immersed in an ice bath. Glass wool inside the sulfur condenser was used to prevent sulfur mist from escaping. The volumetric flow rate of the product gas, free of elemental sulfur, was measured using a Humonics 650 digital soap bubble flowmeter.

Elemental sulfur collected was quantified gravimetrically using an electronic scale with a precision of 0.001 g. The product gas, free of elemental sulfur, was withdrawn from the sampling tube and injected into a gas chromatograph for composition analysis. The pressure drop across the entire reactor system was less than 18 mmHg; thus, the experiments were carried out at atmospheric conditions. Finally, the exit gas passed through a scrubber containing concentrated NaOH solution, and then was vented into a fumehood. The inlet and exit gases were analyzed by using a Varian 3800 gas chromatograph equipped with a 1/4 in. o.d., 6 ft. long Porapak QS column and a thermal conductivity detector (TCD). The detection limit of the GC-TCD is about 0.1 vol % for a 100- μL gas sample. The carrier gas was helium. The GC-TCD was

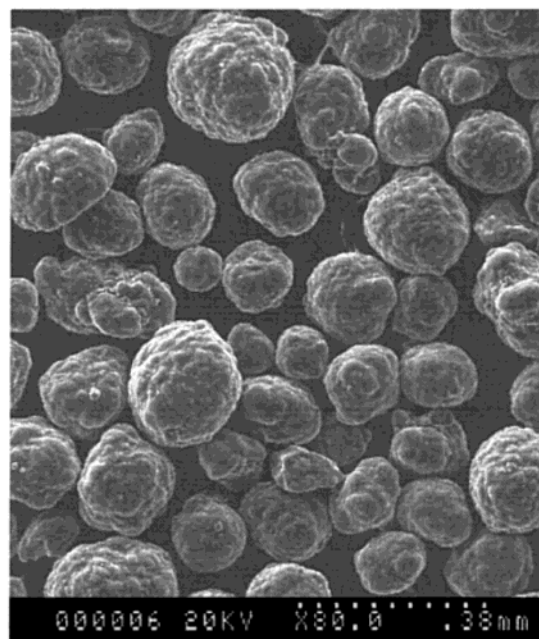


FIGURE 2. SEM image of coke particles.

calibrated for SO_2 , CS_2 , COS, CO_2 , and CO, respectively. The relative standard deviations (RSD) of the GC analyses varied from 0.5 to 10%.

The oilsands fluid coke was obtained from the Syncrude Plant in Alberta, Canada. It is a byproduct from the upgrading process that produces synthetic crude oil out of bitumen. It consists of about 85 wt % carbon, 5% volatiles, 5% silicate-bases ash, and 6–8 wt % sulfur. The sulfur content in the coke used in this study was 8.3 wt % as determined by using a LECO CS244 sulfur analyzer. Physically, the fluid coke was nonporous spherical particles with an “onion like” internal structure. A SEM image of the coke particles is given in Figure 2. The coke was sieved before use, and the size fraction used in the experiments was 205–295 μm . Mass balances of carbon and sulfur species before and after the reaction were within 10% error.

Results and Discussion

Equilibrium Product Composition. The thermodynamic software package F*A*C*T (16), which is based on Gibbs free energy minimization, was used for calculating equilibrium product compositions of C– SO_2 systems at various reactant compositions and temperatures. Calculations were per-

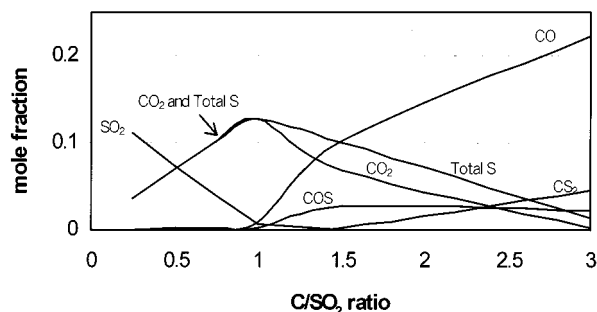


FIGURE 3. Predicted equilibrium product composition as a function of C/SO₂ ratio at 900 °C.

formed for atmospheric systems at 400–1100 °C, with various C/SO₂ ratios. Although F*A*C*T considers all possible products, only the products with molar fraction greater than 1×10^{-5} were presented in this work. They are SO₂, CS₂, COS, CO₂, CO, and elemental sulfur. At the temperatures studied, elemental sulfur, S_x, is in a gaseous form. Although the number of atoms in a molecule, “x”, varies from 1 to 8 depending on temperature, diatomic molecules were found to be predominant. F*A*C*T also predicted the presence of other sulfur oxides, SO and S₂O. The calculated equilibrium SO₂/S₂O partial pressure ratios were always greater than 30 regardless of temperatures. SO was found to be a minor species with equilibrium SO₂/SO ratios from 450 to 47 000, depending on temperatures. In general, lower temperatures favor the formation of SO. Since the system can potentially produce not only elemental sulfur but also COS and CS₂, the determination of conditions for enhancing elemental sulfur production is critical to the development of a coke-based SP-FGD process.

As carbothermal reduction of SO₂ can produce reduced sulfur species other than elemental sulfur, the conversion of SO₂ as well as the yield of elemental sulfur need to be quantified. The elemental sulfur yield (Y_{sulfur}) is defined as

$$Y_{\text{sulfur}} = m_{S_1} / m_{SO_2}^i \quad (1)$$

where, m_{S_1} is the number of moles of elemental sulfur allotropes expressed as S₁, $m_{SO_2}^i$ is the initial number of moles of SO₂. Y_{sulfur} is therefore the ultimate sulfur yield and a measure of selectivity in terms of product type. The fractional SO₂ conversion (X_{SO_2}) is defined as

$$X_{SO_2} = (m_{SO_2}^i - m_{SO_2}^f) / m_{SO_2}^i \quad (2)$$

where $m_{SO_2}^f$ is the number of moles of SO₂ left with final products.

Effect of C/SO₂ Ratio on Equilibria. The effect of C/SO₂ ratio on equilibrium product composition was calculated for systems at 300–900 °C. For all temperatures, the product composition is highly dependent on the C/SO₂ ratio. The product composition as a function of C/SO₂ ratio at 900 °C is shown in Figure 3. Calculations for temperatures below 900 °C gave similar results. An excess of carbon (high C/SO₂ ratio) results in an increase in sulfide species (COS and CS₂), whereas an excess of SO₂ (low C/SO₂ ratio) suppresses the formation of COS and CS₂. Figure 4 shows the predicted SO₂ conversion and elemental sulfur yield as a function of C/SO₂ ratio at 900 °C. As the C/SO₂ ratio increases, the SO₂ conversion increases as expected, and the sulfur yield increases and reaches a maximum at a C/SO₂ ratio of 1, which corresponds to the stoichiometry of the reaction $C + SO_2 = S + CO_2$. A further increase in the C/SO₂ ratio results in a lower elemental sulfur yield due to the reactions between S and C or CO. At C/SO₂ ratios less than 1, as shown in Figure 3, some SO₂ remains unconverted due to the limited

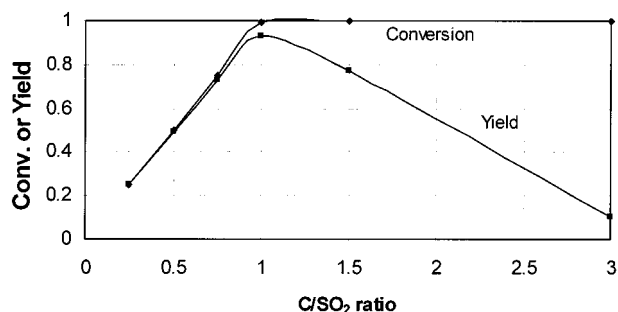


FIGURE 4. Predicted equilibrium SO₂ conversion and sulfur yield as a function of C/SO₂ ratio at 900 °C.

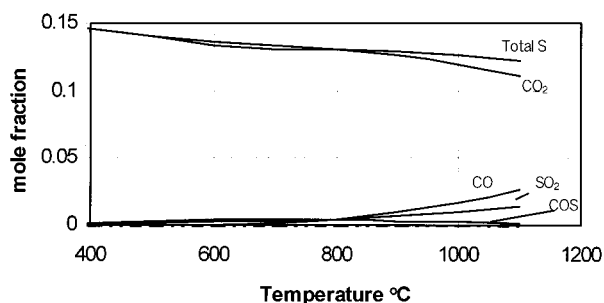
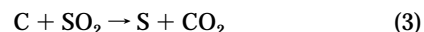


FIGURE 5. Equilibrium product composition as a function of temperature at C/SO₂ = 1.

supply of carbon; CO₂ and elemental sulfur are the major products, indicating that if SO₂ is in excess the predominant reaction is



It should be pointed out that CO₂ is a common species in flue gases. As a major product, CO₂ will inevitably affect the reaction equilibrium. High CO₂ concentrations can potentially limit the reduction of SO₂. F*A*C*T predicted that the equilibrium CO₂/SO₂ ratio increases with the decrease in temperature and the initial C/SO₂ ratio. At 700 °C and C/SO₂ = 1, for example, the equilibrium CO₂/SO₂ ratio was determined to be 55. It reaches about 250 when the C/SO₂ ratio drops to 0.95, still having a SO₂ conversion greater than 90%.

Effect of Temperature on Equilibrium Product Composition. Figure 5 shows the effect of temperature on equilibrium product compositions for systems of unity C/SO₂ ratio. Elemental sulfur and CO₂ were the major products. The amount of elemental sulfur and CO₂ decreased with temperature. At temperatures below 800 °C, the sulfur curve overlaps with the CO₂ curve, again suggesting the stoichiometry of the reaction 3. At temperatures above 800 °C, the C–CO₂ reaction prevails forming CO and resulting in a lower CO₂ mole fraction than elemental sulfur. Higher temperatures inflate the mole fraction of unconverted SO₂ as well as sulfides but to a lesser extent. Figure 6 shows a slight decrease in SO₂ conversion and a significant decrease in elemental sulfur yield as temperature rises. Nevertheless, at temperatures below 1000 °C, the SO₂ conversion and the elemental sulfur yield are still above 90%. This is in agreement with Klinzing and Walker’s (18) simulation of the C–SO₂ system with two thermodynamic models, indicating an ultimate elemental sulfur yield close to 90%.

At C/SO₂ ratios greater than 1, particularly at higher temperatures, product COS and CS₂ become significant. This can be attributed to the increase in the severity of the reducing conditions as temperature and C/SO₂ ratio increase. Figure 7 shows the product composition as a function of temperature for a C/SO₂ ratio of 3. As expected, CO becomes the

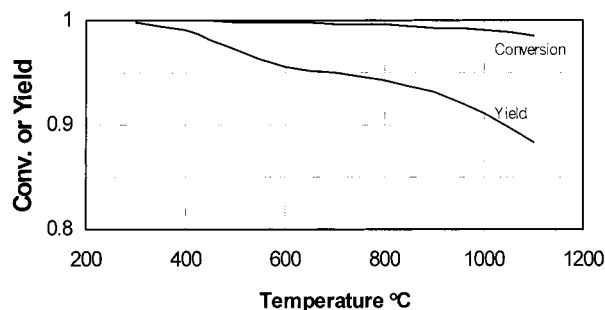


FIGURE 6. Equilibrium SO_2 conversion and sulfur yield as a function of temperature at $\text{C}/\text{SO}_2 = 1$.

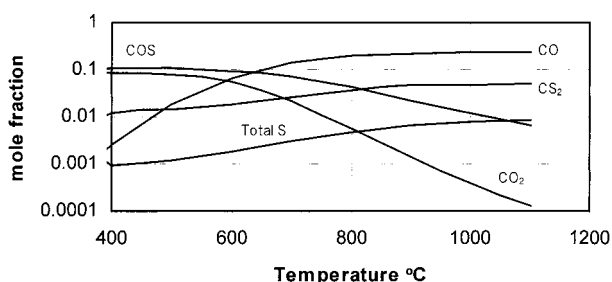


FIGURE 7. Predicted equilibrium product composition as a function of temperature at $\text{C}/\text{SO}_2 = 3$.

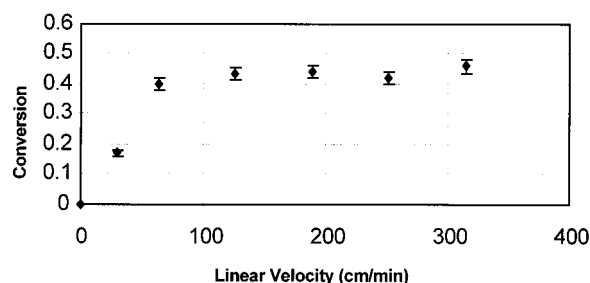


FIGURE 8. SO_2 conversion as a function of linear gas velocity at 700°C and 15 vol % feed SO_2 .

predominant gaseous carbon species at a higher C/SO_2 ratio and high temperatures. At temperatures below 800°C , the predominant sulfur species is COS, whereas CS_2 is the major product at temperature above 800°C . The implication is that the product composition of the reaction (such as elemental sulfur, CS_2 , COS, and CO_2) can be varied by adjusting temperature and relative amounts of C and SO_2 .

Effect of Linear Gas Velocity on SO_2 Conversion. For experimental results, the fractional SO_2 conversion (X_{SO_2}) is defined as

$$X_{\text{SO}_2} = (F_{\text{SO}_2}^i - F_{\text{SO}_2}^o) / F_{\text{SO}_2}^i \quad (4)$$

where, $F_{\text{SO}_2}^i$ is the molar flow rate (mol/min) of feed SO_2 and $F_{\text{SO}_2}^o$ is the molar flow rate of product SO_2 , which is determined by multiplying the outlet volumetric rate (mL/min) by the SO_2 molar concentration (mol/mL).

A series of experiments at 700°C was carried out at various linear gas velocities in order to study the effect of mass transfer on SO_2 conversion. The gas velocity was varied by adjusting the volumetric flow rate of feed gas. To maintain a constant gas residence time in the reactor at various gas velocities, the amount of coke in the reactor, and hence the height of the coke bed, was adjusted accordingly. Figure 8 shows that at low velocities the conversion increased appreciably with velocity. Above 150 cm/min, however, SO_2 conversion became independent of velocity, suggesting an insignificant

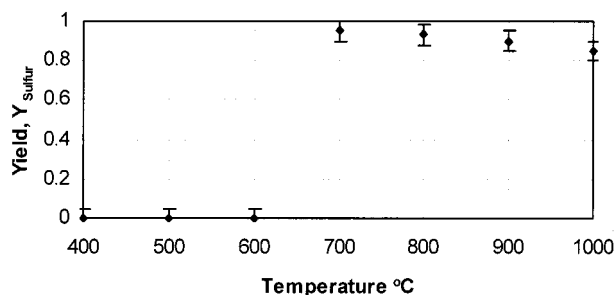


FIGURE 9. Elemental sulfur yield as a function of temperature for 15 vol % feed SO_2 .

effect of mass transfer at high velocity. All the subsequent experiments were performed under the high-velocity flow regime (>150 cm/min).

Temperature Effects on Elemental Sulfur Yield. The yield of elemental sulfur (Y_{sulfur}) is defined as

$$Y_{\text{sulfur}} = (W_s / M_s) / (F_{\text{SO}_2}^i t) \quad (5)$$

where W_s is the weight of product elemental sulfur (g), M_s is the molecular weight of sulfur, and t is reaction time (min).

Figure 9 shows the maximum elemental sulfur yield at various temperatures. At temperatures below 600°C , no elemental sulfur was collected. This result differs from the theoretical prediction (Figure 6) that assumes infinite reaction time. At 700°C , however, the conversion of SO_2 to elemental sulfur was significantly accelerated and reached the maximum ($>95\%$). The hindered reaction kinetics at low temperatures may be a factor but cannot explain the sharpness of the change. The sudden increase in elemental sulfur yield from 600 to 700°C was indeed intriguing, which warrants more detailed investigations at such a temperature range. When temperature increased from 700 to 1000°C , the elemental sulfur yield decreased slightly, but was still above 90% at 900°C . The gradual decrease in sulfur yield with temperature agrees with the theoretical prediction (Figure 6), indicating that the elemental sulfur yield was thermodynamically controlled and reached the equilibrium limit under such conditions. In principle, the amount of sulfur recovered in the condenser by quenching differs from the amount of sulfur in a vapor form at reaction temperatures, if the quenching cannot completely stop the reactions after the gas leaves the coke bed. Since low temperatures favor elemental sulfur over other reduced sulfur, any reaction during quenching will likely result in an overestimation of the amount of sulfur vapor at high temperatures.

Effects of Reaction Time on SO_2 Conversion and Elemental Sulfur Yield. Residence time of the gas in the reactor, τ , is defined as

$$\tau = (V\epsilon) / G \quad (6)$$

where V is the volume (mL) of coke bed, ϵ is the void fraction of the bed, and G is the flow rate (mL/min) of gases to the reactor, measured at standard conditions. The void fraction is determined by

$$\epsilon = (1 - \rho_{\text{ap}} / \rho_{\text{C}}) \quad (7)$$

where ρ_{ap} is the apparent density of the coke (g/mL) and ρ_{C} is the density of carbon (g/mL). Effects of reaction time on SO_2 conversion and elemental sulfur yield were investigated at different temperatures. At a set temperature with 15 vol % feed SO_2 , reaction times were varied by adjusting the flow rate of the feed gas and/or the mass of coke.

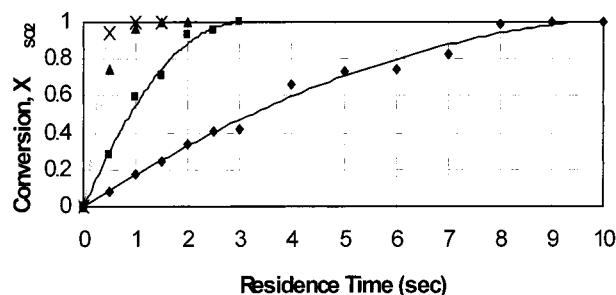


FIGURE 10. SO_2 conversion as a function of time and temperature for 15 vol % feed SO_2 : (♦) 700, (■) 800, (▲) 900, and (×) 1000 °C.

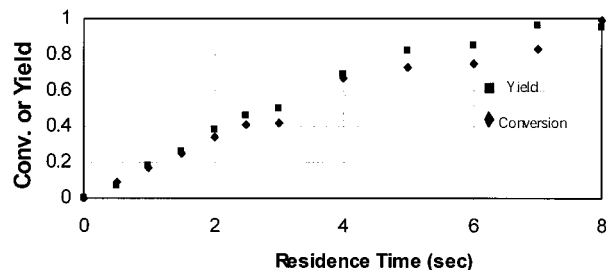


FIGURE 11. SO_2 conversion and elemental sulfur yield as a function of time at 700 °C and 15 vol % feed SO_2 .

Figure 10 shows SO_2 conversion increases with reaction time at various temperatures (700–1000 °C). As temperature increased, the time required for complete conversion of SO_2 decreased. This result is not surprising, since high temperature speeds up the rate of reaction. Figure 11 shows that SO_2 conversion and elemental sulfur yield increased with time at 700 °C. The complete conversion was achieved in about 8 s. At such operating conditions, the amount of SO_2 converted was almost equal to the elemental sulfur yield. Insignificant amounts of COS and CS_2 were detected by the GC-TCD, suggesting that SO_2 was converted to elemental sulfur stoichiometrically following reaction 3, as indicated by the reaction equilibrium calculations. It is worth noting that the flow rate of the product gas after removing elemental sulfur was measured and found to be within 5% of the feed gas rate. This is consistent with the theoretical prediction that reaction 3 is the predominant reaction when the supply of carbon is limited. According to this reaction, the total number of moles of gases should remain constant.

It was also found that the elemental sulfur yields were slightly but consistently higher than the SO_2 conversion. Material balance analysis revealed that the extra elemental sulfur was from the coke that contained about 8 wt % sulfur. Although the sulfur in coke could be removed along with carbon during reaction, the sulfur concentration in the partially reacted coke was found to be higher than that of the original one, suggesting that the rate of carbon consumption by the reaction with SO_2 was higher than the rate of sulfur removal from the coke. The apparent stability of the sulfur in coke is expected given the fact that the coke was produced in a coking reactor at about 600 °C. Figure 12 shows the product compositions as a function of time at 800 °C. No significant reduced sulfur species (COS and CS_2) was detected at short reaction time. But, prolonging gas–coke contacting at high temperature resulted in the significant formation of COS, which is in agreement with theoretical predictions as shown in Figure 5.

In summary, the carbothermal conversion of SO_2 to elemental sulfur using oilsands fluid coke is feasible at temperatures above 700 °C. A complete conversion of SO_2 is readily achievable within seconds. The product distribution can be adjusted by varying process parameters to yield

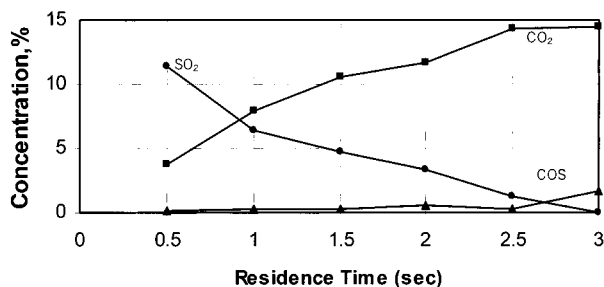


FIGURE 12. Product distribution (elemental sulfur free) as a function of time at 800 °C and 15 vol % feed SO_2 .

elemental sulfur as the sole sulfur product. More work is still needed to examine the influence of other flue gas components such as oxygen and carbon dioxide. Partial combustion may represent an alternative as to how the system would be heated. Maintaining a reducing atmosphere for SO_2 reduction while burning carbon with O_2 can be potentially challenging. But, there are reactors in which the reductive burning is carried out continuously, for example, a blast furnace. A comprehensive kinetic study is also recommended to lay a foundation for the development of a coke-based SP-FGD process. Such a process holds a strong potential to be economically favorable for several reasons: (i) oilsands fluid coke, as the only reagent, is a byproduct that is essentially free; (ii) pretreatment of the coke may not be necessary given its desirable physical properties (e.g., particle size); and (iii) at 700 °C reaction temperature, the cooling of flue gases may be avoided.

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Literature Cited

- (1) Nelli, C. H.; Rochelle, G. T. *J. Air Waste Manage. Assoc.* **1998**, *48*, 819–828.
- (2) Pelley, J. *Environ. Sci. Technol.* **2000**, *34* (11), 248A.
- (3) Livengood, C. D.; Markussen, J. M. *Power Eng.* **1994**, 38–42.
- (4) Abramowitz, H.; Insigna, R.; Rao, Y. K. *Carbon* **1976**, *14*, 84–86.
- (5) INCO Limited, Manitoba Division. *Sulfur control: technical description and analysis of control methods*, 1985.
- (6) Jin, Y.; Yu, Q.; Chang, S. *Environ. Prog.* **1997**, *1* (16), 1–8.
- (7) Biswas, A. K.; Roy, N. C.; Rao, M. N. *Indian J. Technol.* **1968**, *6*, 157.
- (8) Blackwood, J. D.; McCarthy, D. J. *Proc. Australas. Inst. Min. Metall.* **1974**, *249*, 25.
- (9) Steiner, P. J.; Juntgen, H.; Knoblauch, K. *Adv. Chem. Ser.* **1975**, No. 139, 181.
- (10) Foster Wheeler. U.S. Patent 4147762, 1979.
- (11) Ratcliffe, C. T.; Pap, G. *Fuel* **1980**, *59*, 237–243.
- (12) Steiner, P. J.; Gutterman, C.; Dalton, S. M. U.S. Patent 4328201, 1980.
- (13) Jia, C. Q.; Lu, W.-K. *Environ. Sci. Technol.* **1996**, *30* (2), 377–384.
- (14) Lu, W.-K.; et al. U.S. Patent 6030592, 2000.
- (15) Chung, K. H.; Janke, L. C. G.; Dureau, R.; Furimsky, E. *Environm. Sci. Eng.* **1996**, 50–53.
- (16) Furimsky, E. *Fuel Process. Technol.* **1998**, *56*, 263–290.
- (17) Thompson, W. T.; Eriksson, G.; Bale, C. W.; Pelton, A. D. *EQUILIB. In Facility for the Analysis of Chemical Thermodynamics (F*A*C*T), Guide to Operations*; Pelton, A. D., Bale, C. W., Eds.; Thermfact: Montreal, Quebec, 1985.
- (18) Klinzing, G. E.; Walker, R. J. *Fuel* **1984**, *63*, 1450–145.

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