Ca-Mg Acetate as Dry SO_2 Sorbent: III. Sulfation of MgO+CaO

Hong Yong Sohn and Dong Hoon Han
Dept. of Metallurgical Engineering and of Chemical and Fuels Engineering, University of Utah,
Salt Lake City, UT 84112

The separate sulfation rate of MgO contained in calcined calcium magnesium acetate (CMA) by SO_2 and O_2 was measured, as well as the simultaneous sulfation of MgO and CaO in the temperature range where the sulfates of both oxides are stable. In the temperature range below 830°C in which MgO sulfation is thermodynamically possible the reaction produced $CaSO_4 \cdot 3MgSO_4$, rather than $MgSO_4$. The formation of the $CaSO_4 \cdot 3MgSO_4$ phase at a reasonable rate during the sulfation of calcined CMA has a significant implication in that it allows the utilization of MgO for SO_2 removal, whereas the sulfation of MgO from conventional sources is so slow that it is of no practical interest. Thus, the calcined CMA can remove SO_2 at lower temperatures than conventional SO_2 absorbent and at a greater capacity and rate, which presents a substantial competitive advantage of this material.

Introduction

The calcination of calcium magnesium acetate (CMA) produces a mixture of CaO and MgO that has high porosity, large pores, and a large internal surface area (Han and Sohn, 2002). The reaction of the CaO component in calcined CMA above 950°C where only CaO, not MgO, is sulfated was described in Part II of this series (Sohn and Han, 2002). The thermodynamic conditions in which only CaO or both oxides may be sulfated were also examined therein. Part III describes the sulfation reaction involving MgO, separately as well as together with the CaO content in calcined CMA. Information on the separation of the two oxides is required to describe the overall sulfation rate of CaO and MgO in calcined CMA under any conditions, because their sulfation rates have different dependencies on temperature and other conditions.

MgO has a relatively low reactivity towards SO_2, and, thus, it has been considered an inefficient SO_2 absorbent (Borgwardt, 1972; Marier and Dibbs, 1974) and its reactivity has not been much studied. According to the results from this work, MgO from CMA has a much greater potential as a desulfurization reagent. The improvement of the reaction rate and sulfation capacity is due to the formation of the $CaSO_4 \cdot 3MgSO_4$ phase. This fact has an important implication in that calcined CMA can remove SO_2 at lower temperatures than conventional SO_2 absorbent and at a greater capacity.

Experimental Work

Although the sulfation experiment followed the procedure reported in Part II of this series, some important details are repeated here for convenience. A thermogravimetric analysis (TGA) unit consisting of three major parts—an electrobalance, a reactor, and a gas-delivery system—was used in this work (Han and Sohn, 2002). The weight changes taking place during the reaction were measured continuously with a Cahn balance (Model 1000). In the course of the experiment, the balance chamber was purged continuously with nitrogen gas to prevent the intrusion of reaction gas and heat. The furnace tube was an Inconel pipe with 66.0 cm length and 5.1 cm ID. One-fifth of the reactor was filled with ceramic balls for mixing and preheating of the inlet gas. An R type thermocouple (platinum vs. platinum + 13% rhodium) was protected in a 0.6 cm diameter ceramic tubing and positioned adjacent to the sample within the reactor. A sample tray made of alumina, 2.6 cm ID and 0.4 cm depth, was suspended into the uniform temperature zone of the reactor with a platinum wire.

In the determination of intrinsic particle kinetics parameters, it is important to ensure that measurements are carried

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Correspondence concerning this article should be addressed to H. Y. Sohn.

out under the conditions such that diffusion between and within the particles, and external mass transfer, do not affect the overall rate. The particle size of CMA used for this purpose was small and the porosity of calcined CMA was high, as discussed in Part I of this series (Han and Sohn, 2002). The effect of interstitial diffusion was removed by charging a 300 mg sample as a thin layer to minimize the diffusion path. Gas-flow rates in the range of 0.6 to 1.81 NL/min were used to investigate the effect of gas-flow rate (using calcined CMA powder of $-325+400$ mesh in 0.5% SO_2 in the presence of 5% oxygen at 1,000°C). It was determined that the reaction rate was independent of the flow rate above 1.5 NL/min (Han, 1998). Thus, in all the subsequent runs, a gas-flow rate of 1.8 NL/min was chosen so as to avoid any external mass-transfer effects. Drierite dehydration units were placed just before the flowmeters to remove moisture from the gases. Anhydrous grade (minimum 99.9% purity) sulfer dioxide was supplied by Union Carbide. Nitrogen and oxygen were supplied by Air Products Co. and had a minimum 99.9% purity.

The sulfation reaction was carried out under various sulfur dioxide partial pressures and temperatures while keeping the oxygen partial pressure constant at 4.36 kPa.

Experimental Results

Formation of $CaSO_4 \cdot 3MgSO_4$ phase

As discussed earlier, it is not possible to sulfate only MgO in a mixture of CaO and MgO because the equilibrium partial pressure of SO_2 for the sulfation of CaO is much lower than that of MgO (Sohn and Han, 2002). To determine the separate kinetics of MgO sulfation, the calcined CMA samples were reacted under the conditions in which only the CaO was completely converted to $CaSO_4$, and then the temperature of the system was lowered to the desired value where MgO could react with sulfur dioxide. For these runs, the sulfation of CaO was completed at 1,000°C under an SO_2 partial pressure of 0.43 kPa.

The product of MgO sulfation was the $CaSO_4 \cdot 3MgSO_4$ phase hitherto unreported in the sulfation of any materials containing CaO and MgO, rather than separate $MgSO_4$ phase mixed with $CaSO_4$. No pure $MgSO_4$ phase was detected by X-ray, as shown in Figure 1. The reason for the formation of the $CaSO_4 \cdot 3MgSO_4$ phase in this case, but not in other CaO-MgO sorbents, is not certain at this point. The equilibrium phase diagram for the $MgSO_4$-$CaSO_4$ binary system is shown in Figure 2, which was drawn based on the data of Rowe et al. (1967).

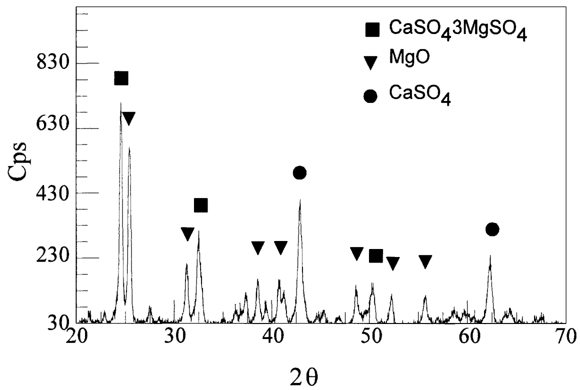

Figure 1. XRD pattern of the MgO sulfation product.

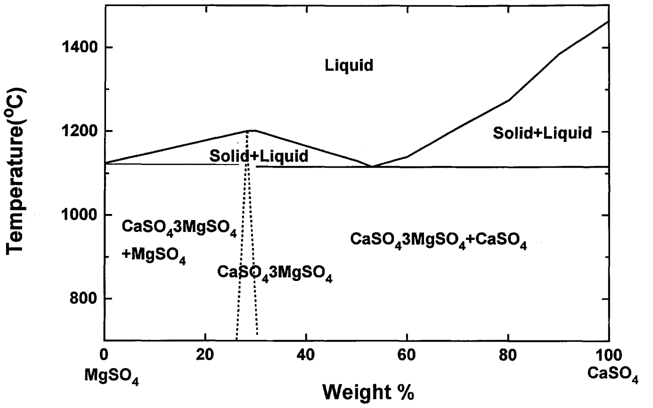

Figure 2. Binary phase diagram for the $CaSO_4$-$MgSO_4$ system.

Intrinsic rate of MgO sulfation

Effect of Reaction Temperature. The effect of temperature on MgO conversion was examined in the range 600 to 830°C, as shown in Figure 3. In these runs, the partial pressures of SO_2 and O_2 were kept at 0.43 and 4.3 kPa (0.5 and 5% by volume under the atmospheric pressure of 86.1 kPa in Salt Lake City), respectively.

The experiment for MgO sulfation was initially carried out from 700 to 830°C under otherwise identical conditions. Contrary to the usual effect of temperature, the reaction rate decreased with increasing temperature in this case, as shown in

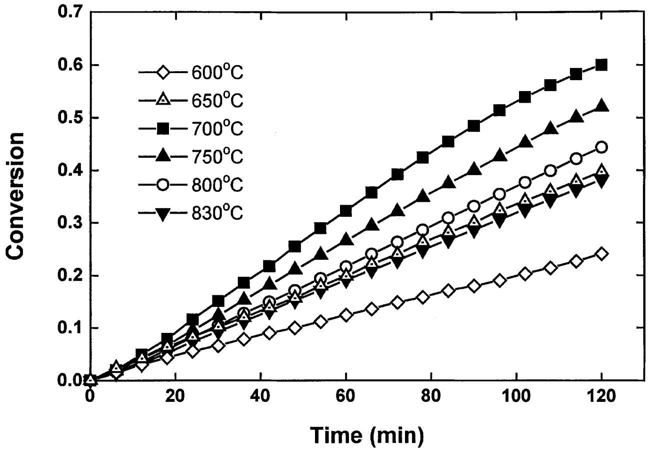

Figure 3. Effect temperature on MgO conversion ($p_{SO_2} = 0.43$ kPa; $p_{O_2} = 4.3$ kPa).

All CaO in the sample was pre-sulfated.

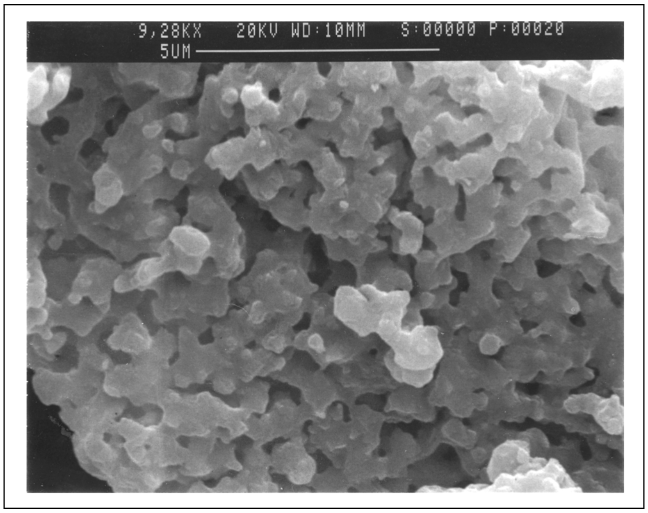

Figure 4. Scanning electron micrograph of calcined CMA after MgO sulfation at 700°C.

Figure 3. Additional experiments showed that the reaction rate increased with the temperature from 600 to 700°C. Scanning electron micrograph (SEM) analysis was conducted to investigate this unusual phenomenon. Figures 4 and 5 show the surface textures of the calcined CMA after the sulfation reaction at 700°C and 830°C, respectively. The micrograph of the sample reacted at 700°C shows that the particle remained porous. However, the grains in the sulfated particle reacted at 830°C shown in Figure 5, although still relatively porous, are covered with agglomerations of small nodules that are welded into significantly larger grains. Among the several possible factors examined based on the results of the microscopic analysis, the most likely reason for this observed temperature dependence was the formation of a $CaSO_4 \cdot 3MgSO_4$ layer the denseness of which increases with the temperature (Han, 1998).

Effect of Sulfur Dioxide Partial Pressure. Figure 6 shows the effect of SO_2 partial pressure on MgO conversion. The SO_2 partial pressure was varied between 0.26 and 0.86 kPa (0.3 and 1 vol. %). The temperature of the system and the partial pressure of the oxygen were kept constant at 750°C and 4.3 kPa, respectively.

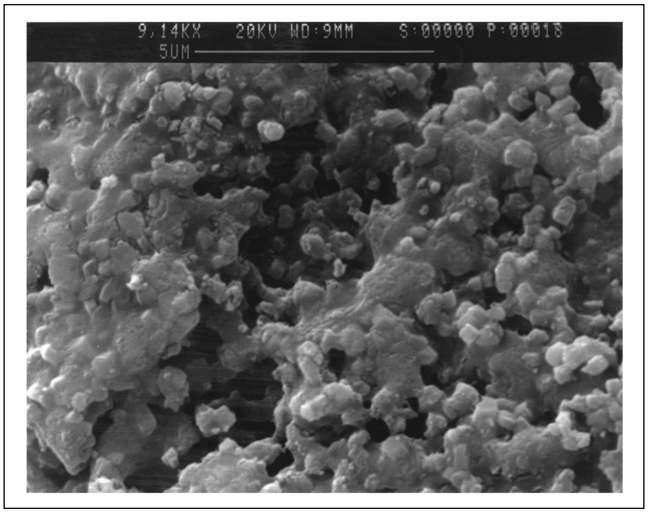

Figure 5. Scanning electron micrograph of calcined CMA after MgO sulfation at 830°C.

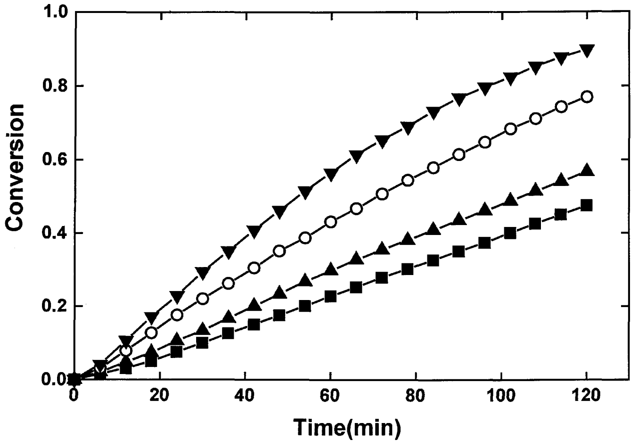

Figure 6. Effect of SO_2 partial pressure on MgO conversion ($p_{O_2} = 4.3$ kPa; T = 750°C).

All CaO was pre-sulfated. ■ 0.26 kPa; ▲ 0.43 kPa; ○ 0.61 kPa; ▼ 0.86 kPa.

Attainment of high degrees of conversion

As stated in Part II of this series, typical sulfation rates level off as the reaction proceeds due to the formation of a dense product layer (Chang and Thodos, 1984; Hajaligol et al., 1988; Kim, 1999), even when the reactant oxide has some initial porosity. It is, therefore, of great interest to note that the sulfation of MgO in the calcined CMA, like that of CaO in the same material (Sohn and Han, 2002), does not level off even at high conversion levels. This has an important implication in that all of the calcined CMA can be utilized for SO_2 removal, thus, making its sulfation capacity very high. The reason for this behavior was explained in Part II of this series.

Kinetics of MgO Sulfation

The nucleation-and-growth kinetics model, which describes the sulfation of CaO in calcined CMA, is also applicable for the analysis of kinetics of MgO sulfation. The rate of MgO sulfation was, thus, analyzed using the following rate equation

$$[-\ln(1-\alpha)]^{1/m} = k_{app} \cdot t \tag{1}$$

where α is the fractional conversion, m is a constant, and k_{app} is the apparent rate constant which is given by

$$k_{app} = k \cdot f(p_{SO_2}) \tag{2}$$

where k is the rate constant, p_{SO_2} is the sulfur dioxide partial pressure, and f designates the partial pressure dependence of rate. The rate constant k is related to temperature by

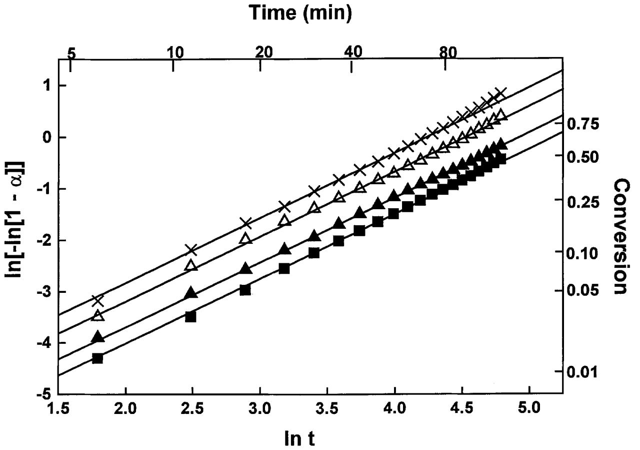

Figure 7. Plot of ln[−ln(1 − α)] vs. ln (t) according to Eq. 1 from the results in Figure 6.

■ 0.26 kPa; ▲ 0.43 kPa; △ 0.61 kPa; × 0.86 kPa.

$$k = k_o \cdot \exp\left(-\frac{E}{RT}\right) \tag{3}$$

where k_o is the pre-exponential factor, E is activation energy, R is gas constant, and T is absolute temperature. It is apparent from Eq. 1, that a plot of $\ln[-\ln(1-\alpha)]$ vs. $\ln(t)$ should be linear with m as the slope and $m\ \ln(k_{app})$ as the intercept with $\ln(t) = 0$.

The data shown in Figure 6 were plotted according to Eq. 1 to determine the effect of SO_2 partial pressure on MgO sulfation rate. Straight lines through the data are obtained, as shown in Figure 7, and the m values obtained by linear regression analysis were between 1.24 and 1.28 (an average of 1.26). The values of $k_{app,2}$ obtained from the intercepts are plotted in Figure 8 against the sulfur dioxide partial pressure. An overall linear relationship going through the origin is observed, indicating that MgO sulfation rate is of the first order with respect to SO_2 partial pressure. Based on Eq. 2, the slope of the straight line in Figure 8 corresponds to the rate constant k at that temperature.

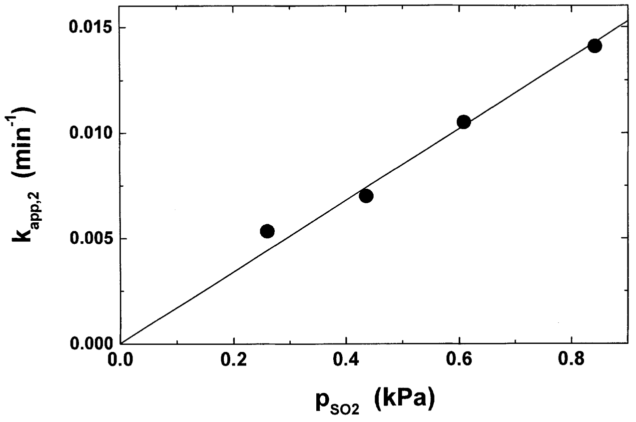

Figure 8. Dependence of the apparent rate constant on SO_2 partial pressure for MgO sulfation.

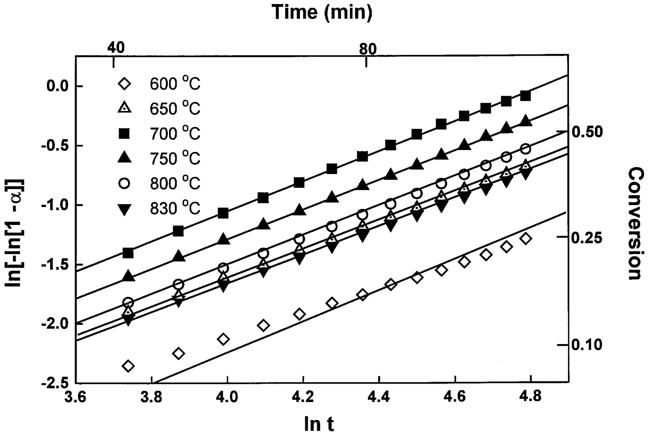

Figure 9. Plot of ln[−ln(1 − α)] vs. ln (t) according to Eq. 1 from the results in Figure 3.

Figure 9 shows a plot of $\ln[-\ln(1-\alpha)]$ vs. ln t obtained from the results of Figure 3 using $m = 1.26$ from the above. It is seen that excellent straight lines are indicated at all temperatures except the lowest temperature of 600°C at which the reaction rate and conversion are slow. The values of k at different temperatures were obtained, as described above, from the intercepts in Figure 9 together with $f(p_{SO_2}) = p_{SO_2}$ in Eq. 2. Combining all the above information, the equation for the intrinsic kinetics, unaffected by mass and heat transfer, of the MgO sulfation is, thus, represented by

$$[-\ln(1-\alpha)]^{1/1.26} = k_{app} \cdot t \tag{4}$$

where $k_{app} = k_2 \cdot p_{SO_2}$, and k_2 is shown in Figure 10. As discussed in Part II of this series, oxygen partial pressure does not affect the rate in a typical range encountered in a dry desulfurization process (Simons et al., 1988), and, thus, does not appear in the rate equation. There were only three temperature values tested in the range where the rate increased with temperature, and, thus, determination of the activation energy obtained from them may be risky. However, the three corresponding values of k_2 formed a straight line on an Arrhenius plot and gave an activation energy of 57.8 kJ/mol in the temperature range 600–700°C.

Simultaneous Sulfation of CaO and MgO

There have been no reported investigations dealing with the simultaneous reaction of CaO and MgO with SO_2 in the presence of oxygen because the reactivity of MgO with SO_2 for desulfurization with conventional agents is very low (Borgwardt, 1972; Marier and Dibbs, 1974). In the case of calcined CMA, however, the sulfation rate of MgO is greatly enhanced due to the formation of the $CaSO_4 \cdot 3MgSO_4$ phase, as discussed earlier. The kinetics of the simultaneous sulfation of CaO and MgO in calcined CMA were determined in this work by the same experimental technique as that of CaO or MgO. In all experiments, calcined CMA was reacted with sulfur dioxide in the presence of oxygen in the temperature range where both MgO and CaO are sulfated (Sohn and Han, 2002).

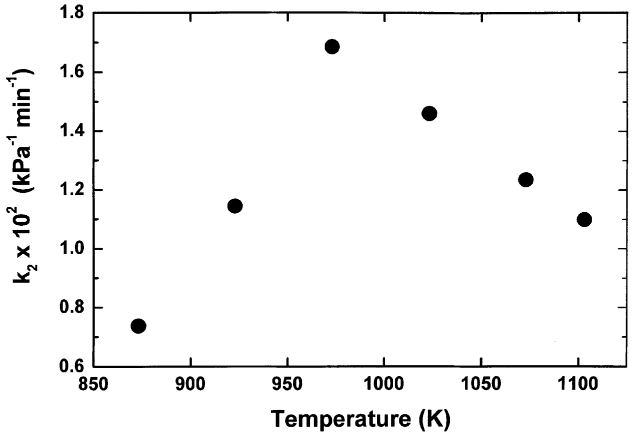

Figure 10. Relationship between rate constant k_2 and temperature for MgO sulfation.

Formulation of overall rate equation

The rate of MgO sulfation analyzed above was obtained in the presence of $CaSO_4$ to form $CaSO_4 \cdot 3MgSO_4$. Thus, in order for that rate expression for MgO sulfation to be applicable to the formulation of an overall rate equation for the simultaneous sulfation of CaO and MgO, a sufficient amount of CaO must be present and its sulfation rate must be sufficiently high to ensure the presence of $CaSO_4$ for MgO sulfation. The first part of this condition was satisfied in this work, because the molar ratio CaO/MgO of 3/7 in the calcined CMA is greater than the ratio of 1/3 in $CaSO_4 \cdot 3MgSO_4$. The second part of the condition was examined by comparing the rate of formation of $CaSO_4 \cdot 3MgSO_4$ from a $CaSO_4$/MgO mixture with the sulfation rate of CaO in a mixture with MgO. The latter was obtained by extrapolating the rate equation for CaO sulfation down to 700°C, because the direct measurement of the sulfation rate of just CaO at this temperature is not possible as MgO is simultaneously sulfated. The extrapolated sulfation rate of CaO is much faster than the rate of MgO sulfation, especially considering that the initial amount of CaO is in excess of 1/3 the amount of MgO necessary to form $CaSO_4 \cdot 3MgSO_4$ (Han, 1998).

When calcined CMA is reacted with SO_2, the overall reaction rate is expressed by

$$\frac{d\alpha_{OV}}{dt} = \frac{d\,{}^{\Delta W_{ac}}/\Delta W_{\text{th}}}{dt} \tag{5}$$

where ΔW_{ac} is the actual combined weight change at any time due to the sulfation of some of the CaO to $CaSO_4$ and some of the MgO to $MgSO_4$ in $CaSO_4 \cdot 3MgSO_4$, and ΔW_{th} is the theoretical weight change for complete sulfation of both CaO and MgO in the calcined CMA. As shown in Part II of this series and above, these are the only sulfation products of CaO and MgO in calcined CMA. Thus

$$\frac{d\alpha_{OV}}{dt} = \frac{1}{\Delta W_{\text{th}}} \cdot \frac{d\Delta W_{ac}}{dt} = \frac{1}{\Delta W_{th}} \cdot \frac{d(\Delta W_{\text{CaO}} + \Delta W_{\text{MgO}})}{\text{dt}} \tag{6}$$

Upon rearranging

$$\frac{d\alpha_{OV}}{dt} = \frac{\Delta W_{\text{CaO}}^{\text{th}}}{\Delta W_{\text{th}}} \cdot \frac{d\alpha_{\text{CaO}}}{dt} + \frac{\Delta W_{\text{MgO}}^{\text{th}}}{\Delta W_{\text{th}}} \cdot \frac{d\alpha_{\text{MgO}}}{dt} \tag{7}$$

where $\Delta W_{\text{CaO}}^{\text{th}}$ is the theoretical weight change for CaO sulfation, and $\Delta W_{\text{MgO}}^{\text{th}}$ is that for MgO sulfation. The values of $\Delta W_{\text{CaO}}^{\text{th}}/\Delta W_{\text{th}}$ and $\Delta W_{\text{MgO}}^{\text{th}}/\Delta W_{\text{th}}$ are 0.3 and 0.7, respectively, from the molar ratio of CaO to MgO in the sample, and thus

$$\frac{d\alpha_{OV}}{dt} = 0.3 \cdot \frac{d\alpha_{\text{CaO}}}{dt} + 0.7 \cdot \frac{d\alpha_{\text{MgO}}}{dt} \tag{8}$$

The conversion rate of CaO on the righthand side is obtained as a function of α_{CaO} by differentiating the following conversion-vs-time relationship given in Part II of this series.

$$[-\ln(1-\alpha_{\text{CaO}})]^{1/1.65} = k_{\text{app}} \cdot t \tag{9}$$

where

$$k_{\text{app}} = 3.26 p_{SO_2} \exp(-E/RT) \text{ min}^{-1} \tag{10}$$

and $E = 37.7$ kJ/mol and p_{SO_2} is in kPa. Under the simultaneous presence of $CaSO_4$, assured as described above, the conversion rate of MgO in Eq. 8 is obtained as a function of α_{MgO} by differentiating Eq. 4. It is noted that integration of Eq. 8 yields

$$\alpha_{\text{OV}} = 0.3\alpha_{\text{CaO}} + 0.7\alpha_{\text{MgO}} \tag{11}$$

with α_{CaO} and α_{MgO} obtained as functions of time from Eqs. 9 and 4, respectively. When the conditions vary with time during the sulfation reaction, the differential form of the rate equation given by Eq. 8 needs to be used.

Rate measurement and comparison with rate equation

CMA particles were charged in a sample tray as a thin layer and calcined completely by heating them at 10°C/min up to 1,000°C, and then the temperature was lowered to the experimental temperature for sulfation. When the temperature became stable, the reactant gases were introduced.

Effect of Sulfur Dioxide Partial Pressure. The partial pressure of sulfur dioxide was varied from 0.26 (0.3 vol. %) to 0.86 kPa (1 vol. %). Temperature and oxygen partial pressure were kept at 750°C and 4.3 kPa, respectively. The effect of sulfur dioxide on the sulfation rate is shown in Figure 11 under otherwise identical conditions. Also shown in this figure are the simulation lines obtained by applying Eq. 8 or 11, indicating close agreement with the experimental results.

Effect of Reaction Temperature. The influence of temperature on the simultaneous sulfation of CaO and MgO was examined in the range 700–830°C, as shown in Figure 12. In these runs, the SO_2 and O_2 partial pressures were kept constant at 0.43 and 4.3 kPa, respectively. It is seen that the reaction rates remain almost the same at different temperatures until the conversion reaches around 0.4, before decreas-

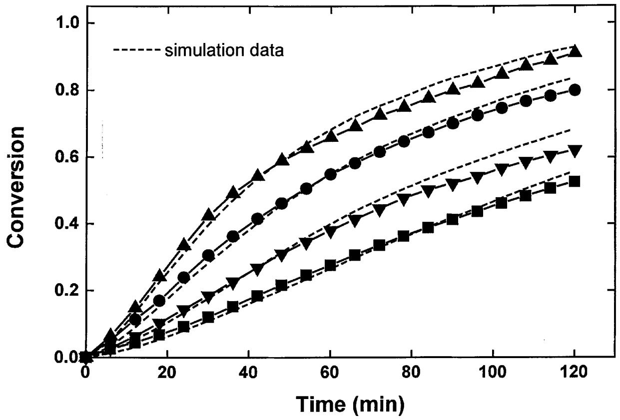

Figure 11. Experimental and simulated results for the effect of SO_2 partial pressure on the simultaneous sulfation of CaO and MgO ($p_{O_2} = 4.3$ kPa; $T = 750°C$).

Conversion here represents the overall uptake of SO_2 relative to the amount required for the complete sulfation of both CaO and MgO in the sample. ■ 0.26 kPa; ▼ 0.43 kPa; ● 0.61 kPa; ▲ 0.86 kPa.

ing with increasing temperature. This trend is due to the different temperature dependencies of the sulfation kinetics of CaO and MgO in calcined CMA. As discussed above, the rate of CaO sulfation increases with temperature. On the other hand, the rate of MgO sulfation decreases with increasing temperature beyond 700°C. In the early stage of the reaction, the overall sulfation rate does not vary much with the temperature because the two reactions have opposite temperature dependencies. As the reaction proceeds, CaO is converted completely before MgO, and, thus, in the second stage of the reaction, the MgO sulfation reaction determines the overall rate.

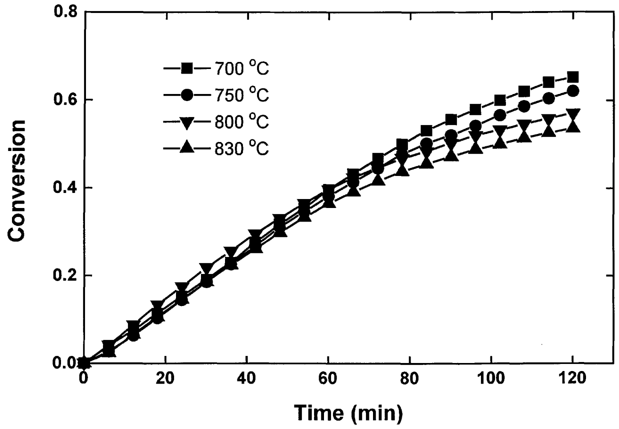

Figure 12. Experimental results for the effect of temperature on the simultaneous sulfation of CaO and MgO ($p_{SO_2} = 0.43$ kPa; $p_{O_2} = 4.3$ kPa).

Conversion here represents the overall uptake of SO_2 relative to the amount required for the complete sulfation of both CaO and MgO in the sample.

The formation rate equation given by Eq. 8 was also applied to the results obtained under different temperatures in this work. Although not shown here, the predictions were in all cases in agreement with the experimental results to the same degree as with the results under various sulfur dioxide partial pressures discussed above and shown in Figure 11 (Han, 1998).

Comparison of SO_2 Removal Data from Various Sources

The reactivities of CaO samples from various sources, some of which contain MgO, were compared with those of calcined CMA. The comparison was made at 900 and 750°C. The partial pressure of sulfur dioxide was fixed at 0.26 kPa. As discussed above, only CaO is expected to react with SO_2 at 900°C, while both CaO and MgO are expected to be sulfated at 750°C. The source materials compared were CMA, Tymochtee dolomite, Greer limestone, Iceland spar limestone, Fredonia limestone, and Rigsby limestone. The data for materials other than CMA were obtained from the literature (Chang and Thodos, 1984; Hajaligol et al., 1988). As shown in Figure 13, the sulfation rate of the calcined CMA is much higher than those of other materials. Furthermore, the calcined CMA absorbs a much greater amount of SO_2 based on the unit amount of CaO than other sources. At 750°C, the capacity of SO_2 absorption per unit kg of CaO by CMA increases from 1.14 to 3.80 kg, because MgO in the calcined CMA participates in sulfation reaction by forming $CaSO_4 \cdot 3MgSO_4$, as discussed earlier. Thus, calcined CMA absorbs a greater amount of sulfur dioxide more rapidly at a lower temperature than other conventional desulfurization agents.

Concluding Remarks

The reactivity of calcined calcium magnesium acetate (CMA) as an SO_2 absorbent has been investigated. Chevron ICE-B-GON Deicer with an atomic ratio of Ca to Mg of 3 to 7 and particle size of $-325+400$ mesh was used as the sample.

Separate sulfation kinetics of MgO were determined in this work, and the following important results were obtained:

(1) The sulfation rate of MgO in calcined CMA is much higher than that contained in other desulfurization agents like calcined dolomite, most likely because of the large internal surface area and the formation of $CaSO_4 \cdot 3MgSO_4$ phase. This also greatly enhances the sulfation capacity of the calcined CMA by allowing the utilization of the MgO content in desulfurization, unlike in the case of other MgO-containing materials.

(2) The sulfation rate of MgO in calcined CMA remains relatively undiminished up to high levels of conversion (unlike those of other desulfurization agents), which results in its high utilization efficiency. This is because the pore size and porosity of calcined CMA are much larger than those of other materials, thus, minimizing the effect of a solid volume increase upon sulfation. This is an important factor that contributes to the high desulfurization capacity of calcined CMA.

(3) A further advantage of the simultaneous sulfation of MgO and CaO in calcined CMA is that the desulfurization can be performed at lower temperatures than with other conventional agents.

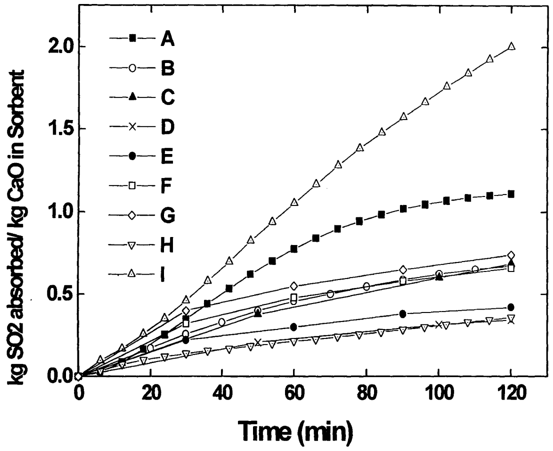

Figure 13. Comparison of the reactivity of CaO obtained from various sources.

p_{SO_2} = 0.26 kPa; P_{O_2} = 4.3 kPa. For T = 900°C: A = CMA, B = Ca(OH)$_2$, C = Tymochtee dolomite, D = Greer limestone, E = Iceland spar limestone, F = Rigsby limestone, G = Fredonia limestone. For T = 750°C: H = Ca(OH)$_2$, I = CMA. (A, B, H and I from this work; C and D from Chang and Thodos [1984]; E, F, and G from Hajaligol et al. (1988)).

A pseudo-first-order kinetics expression is applicable for the sulfation of magnesium oxide. The reaction is of first order with respect to sulfur dioxide partial pressure with an approximate activation energy value of 58 kJ/mol between 600–700°C. At higher temperatures, the rate of this reaction decreases with temperature for reasons discussed above.

The rate of simultaneous sulfation of CaO and MgO in calcined CMA does not vary much with the temperature, because their sulfation rates have opposite temperature dependencies in the tested range 700–830°C. The overall sulfation rate can be closely predicted by an equation formulated based on the kinetics of separate sulfation reactions of the two oxides.

Acknowledgment

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Literature Cited

Borgwardt, R. H., "Properties of Carbonate Rocks Related to SO$_2$ Reactivity," *Environ. Sci. Technol.*, **6**, 350 (1972).

Chang, E. Y., and G. Thodos, "Complex Nature of the Sulfation Reaction of Limestone and Dolomite," *AIChE J.*, **30**, 450 (1984).

Hajaligol, M. R., J. P. Longwell, and A. F. Sarofim, "Analysis and Modeling of the Direct Sulfation of CaCO$_3$," *Ind. Eng. Chem. Res.*, **27**, 2203 (1988).

Han, D. H., "Calcined Calcium Magnesium Acetate (CMA) as a New High-Performance Sulfur Dioxide Absorbent," PhD Diss., Univeristy of Utah, Salt Lake City, UT (1998).

Han, D. H., and H. Y. Sohn, "Calcined Calcium Magnesium Acetate as a Superior SO$_2$ Sorbent: I. Thermal Decomposition," *AIChE J.*, **48**, 2971 (2002).

Kim, B.-S., "Reduction of Sulfur Dioxide to Elemental Sulfur by a Cyclic Process Involving Calcium Sulfide and Sulfate," PhD Diss., University of Utah, Salt Lake City, UT (1999).

Marier, P., and H. P. Dibbs, "The Catalytic Conversion of SO$_2$ to SO$_3$ by Fly Ash and the Capture of SO$_2$ and SO$_3$ by CaO and MgO," *Thermochimica Acta*, **8**, 155 (1974).

Rowe, J. J., G. W. Morey, and C. C. Silber, "The Ternary System K$_2$SO$_4$-MgSO$_4$-CaSO$_4$," *J. of Inorg. Nucl. Chem.*, **29**, 925 (1967).

Simons, G. A., T. E. Parker, and J. R. Morency, "The Oxygen Reaction Order of SO$_2$ with CaO," *Combust. and Flame*, **74**, 107 (1988).

Sohn, H. Y., and D. H. Han, "Ca-Mg Acetate as Dry SO$_2$ Sorbent: Part II. Sulfation of CaO in Calcination Product," *AIChE J.*, **48**, 2978 (2002).

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