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REGENERATIVE PROCESS FOR DESULFURIZATION OF HIGH TEMPERATURE COMBUSTION AND FUEL GASES

QUARTERLY PROGRESS REPORT NO. 11
OCTOBER 1 - DECEMBER 31, 1978

MASTER

PROCESS SCIENCES DIVISION
DEPARTMENT OF ENERGY AND ENVIRONMENT

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REGENERATIVE PROCESS FOR DESULFURIZATION OF HIGH TEMPERATURE COMBUSTION AND FUEL GASES

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Summary

In the continuing search for more reactive calcium silicates, Portland cement, as an ingredient of concrete, has been identified to have favorable properties for sulfur absorption and regeneration. Various Portland cements have been studied for their reactivities for sulfation. Most of Portland cements showed equal or higher sorption reactivities than lime. Portland Type III had unusually high activity for a calcium-based sorbent. Furthermore, pellets could be formed from Portland Type III which had high resistance to attrition. Cyclic sulfation and regeneration using Portland Type III have been conducted in a TG system and there was no sign of weakening of the SO_2 sorption activity. The increased reactivity due to increased porosity of Portland cement pellet was noted by the addition of burnable carbon. The effects of both catalysis and porosity can be seen using fly ash as an additive.

Sulfation and attrition tests for pellets of limestone fines with various binders have been tested. The results showed that both the strength and the sulfation reactivity were higher for pellets of the limestone fines with colloidal silica binder than for raw limestone.

The effects of calcination temperature on the development of pore size and on the SO_2 sorption capacity have been investigated. It has been shown that the temperature maximum observed in a FBC can very possibly be due to pore size variations because of calcinations at different temperatures.

A low-temperature solid state process of forming fine particles of reactive β-dicalcium silicate by firing a mixture of calcium sulfate and silica together with calcium sulfide as a reducing additive is being studied. The temperature of the process can be as low as 850°C. This is expected to result in substantial energy savings and to yield more reactive products.

A rotary kiln, 3 inch ID, is being constructed with the features of external heating and continuous solid feeding. Fabrication of the rotary kiln will be completed during the next report period. It will be in operation for gathering kinetic and process development data. The regeneration of limestones, calcium silicates and Portland cements using fly ash from FBC will be tested with this device.

A. Portland Cement and Calcium Silicates as Regenerative Sorbents

(M. Shen, G. Farber and J. Pruzansky)

Portland cement, as an ingredient of concrete, is a construction material. It may be defined as a product obtained by intimately mixing together calcareous and argillaceous, or other silica, alumina, and iron oxide-bearing materials, burning them at a clinkering temperature (approximately 1400°C), and grinding the resulting clinker.[1] In the continuing search for new and better regenerative SO_2 sorbents for fluidized-bed combustion, we have found that Portland cement has good potential. It has both superior sorbing and regeneration ability and remains strong at high temperatures. A great variety of Portland cements are used throughout the world. Analyses[1] of some American Portland cements are given in Table A-1.

Initial screening and cycling was performed on fine powders. As the program progressed and our efforts centered on the most promising candidates, experimentation shifted to the fabrication and evaluation of pellets. The rate of sulfation of four types of Portland cement are compared with that of aluminous cement in Figure A-1. The reaction conditions were similar to those encountered in fluidized-bed combustion. The sulfation gas contained 0.25% SO_2, 5% O_2, and 15% CO_2 with the balance being N_2. Unlike limestone, Portland cement did not carbonate in a 15% CO_2 atmosphere. Portland Type III, which is intrinsically most reactive to SO_2, is a rapid-hardening cement, similar to ordinary Portland cement type I, except Type III is normally ground finer with slight variations in composition. Type II is slightly slower in rate of strength development than Type I but has additional limitations on the chemical composition. Type V, corresponding to sulphate-resisting Portland cement, is a material with a composition so adjusted as to give it an increased resistance to sulphate-bearing waters.[1] Aluminous cement has the lowest sulfation reactivity due to its low content of calcium oxide (36-40%).

Portland cement Type III powders were subjected to sulfation-regeneration cycling in a TGA. Cycling studies on new materials were an effective way of weeding out impractical substances. In general, sulfation was undertaken at 900°C using 0.25% SO_2, 5% O_2, and 15% CO_2 with the balance being N_2 at a total pressure of one atmosphere. The sulfation period utilized was 2 hours. Regeneration was performed using a simulated environment of 5% CO and 20% CO_2 in nitrogen at 1000°C. When regeneration was completed, before sulfation began, about 5% O_2, 15% CO_2 and balance N_2 was added until no weight gain was observed (the reduced metals were oxidized).

The cycling results obtained are presented in Figure A-2. This figure shows the sulfation level remaining after regeneration for each cycle. The extent of sulfation of the second, third and fourth cycle decreased due to incomplete regeneration of the previous cycle. After the fourth cycle the subsequent three cycles showed no further loss. It is thought that SO_2 has to diffuse through the unregenerated $CaSO_4$ product layer, thereby decreasing the sulfation rate. Portland Type III contains about 2% free lime. It forms $CaSO_4$ upon sulfation, and is therefore more difficult to regenerate than sulfated silicate or aluminate. An attractive property of Portland cement is that the thermodynamics for regenerating these sorbents is much more favorable than for limestone, i.e., higher SO_2 levels could be produced at lower temperatures. Indeed, SO_2 partial pressures at equilibrium when regenerating these sorbents is so high that, unlike the case with limestone, thermodynamics will not be the limiting factor in determining SO_2 level.

After cycling studies, we then focused our efforts on preparing pellets from only those sorbents with the best sulfation/regeneration characteristics. Because of its wide commercial availability Portland Type III cement pellets of a size which could be used in a fluidized-bed combustor were prepared for test purposes. Future work will be conducted in cycling studies using other types of Portland cement to experimentally determine which has the best overall sulfation/regeneration characteristics including the consideration of the effect of free lime.

We found that highly attrition resistant pellets, with good reactivity, could be made from Portland cements. Our goal was to produce pellets with high resistance to attrition and acceptable activity for use in a fluidized bed. The first Portland cement pellets were made from Portland Type III powder, mixed with minimum amount of water, cured, and were subsequently broken and sieved to 16/20 mesh. Pellets extruded through a glass tube were made by mixing the cement powder with a minimum amount of water until a paste formed. The resulting pellets were then humidified for about 28 days to promote proper curing. Pellets averaging 1 mm in length were cut from the extruded strip, dried, and heated to promote hardening.

The extruded materials were subjected to sulfation and regeneration cycling using the TGA to test sorbent reactivity and maintenance of reactivity with cycling. The results of ten cycles are shown in Figure A-3. After the third cycle, the reactivity of the sorbent did not decay any further.

Laboratory experiments confirmed that the activity of β-dicalcium silicate can be improved further through the use of additives. As shown in Figure A-4, by mixing 10% Illinois No. 6 coal ash (< 400 mesh) with β-dicalcium silicate, the reactivity of an extruded pellet was increased by about 50%. The mineral impurities in coal ash probably catalyze the sorption of SO_2 by β-dicalcium silicate. A similar effect was observed in Portland cement. As shown in Figure A-5, the utilization, or fraction converted to sulfate, of extruded pellets prepared with equal weights of Portland Type III and coal ash were about 30% over the utilization of Portland Type III prepared without coal ash. The increased reactivity due to increased porosity of Portland cement pellets were also noted by the use of "burnables". Addition of small amounts of carbon to the cement which is subsequently burned out leaves a porous pellet structure. It has been reported by Hartman and Coughlin[2] that most sulfation takes place in larger pores ($\geq$ 0.4 um) and that pores smaller than 0.4 um are relatively easy to plug. The influence of porosity on the reactivity of Portland cement is obvious. Figure A-5 also shows that by adding one percent cocoanut charcoal to the Portland Type III, the utilization of the extruded pellet increased significantly. The effects of both catalysis and porosity can be seen using fly ash as an additive. For example, ANL LST-7B 2nd cyclone fly ash contained 8.55% carbon. We mixed Portland Type III with an equal weight of ANL LST-7B 2nd cyclone fly ash, and obtained the results as shown in Figure A-6. Because of the weight of the inert materials, the sulfation rate can also be compared to the mass pickup of SO_3 per unit mass of sorbent. Figure A-6 shows the sulfation result based on per unit mass. A comparison of sulfation rates show they are about the same, i.e., the inexpensive inert fly ash may be considered to be equivalent to CaO.

It was thought for economic considerations, and possible structural value cement clinkers could advantageously be used as sorbent pellets. However, the cement clinkers, due to their lack of porosity, are found to be less reactive. Work on experimental techniques to create clinker porosity is necessary.

TABLE A-1
Composition of some American Portland cements

ASTM type	CaO	MgO	Al_2O_3	Fe_2O_3	SiO_2	TiO_2	Na_2O	K_2O	SO_3	Free CaO	C_4AF	C_3A	C_3S	C_2S
I	63.8	3.7	5.6	2.4	20.7	0.23	0.21	0.51	1.5	0.4	7	11	55	18
	63.1	2.5	4.7	3.0	22.1	0.21	0.06	1.30	1.7	0.2	9	7	47	28
	65.8	1.1	4.7	2.1	22.2	0.30	0.04	0.19	1.6	1.6	6	9	54	23
	62.8	1.7	6.7	2.5	21.1	0.39	0.95	0.51	1.8	2.0	8	14	33	35
II	61.4	3.1	4.8	4.8	20.8	0.21	0.06	1.30	1.8	0.9	15	5	44	26
	64.9	1.9	4.0	2.1	24.0	0.23	0.23	0.55	1.7	1.5	6	7	41	38
III	65.6	1.4	5.2	2.5	20.0	0.27	0.21	0.44	2.3	1.8	8	10	63	10
	63.3	4.3	5.1	2.0	20.3	0.21	0.19	0.28	2.5	1.9	6	10	51	19
IV	59.6	3.0	4.5	5.0	22.9	0.23	0.06	1.19	1.3	0.4	15	4	25	47
	63.6	1.1	3.7	3.1	25.2	0.19	0.33	0.01	1.9	0.4	9	5	31	49
V	64.3	1.7	3.1	3.3	24.4	0.19	0.08	0.22	1.4	0.5	10	3	45	36
	64.2	2.5	1.9	1.3	26.1	0.12	0.10	0.15	2.0	1.8	4	3	35	48
	63.3*	1.2	3.3	4.7	23.1	——	0.08	0.37	1.7	——	14	1	49	30

*Corrected for free CaO.

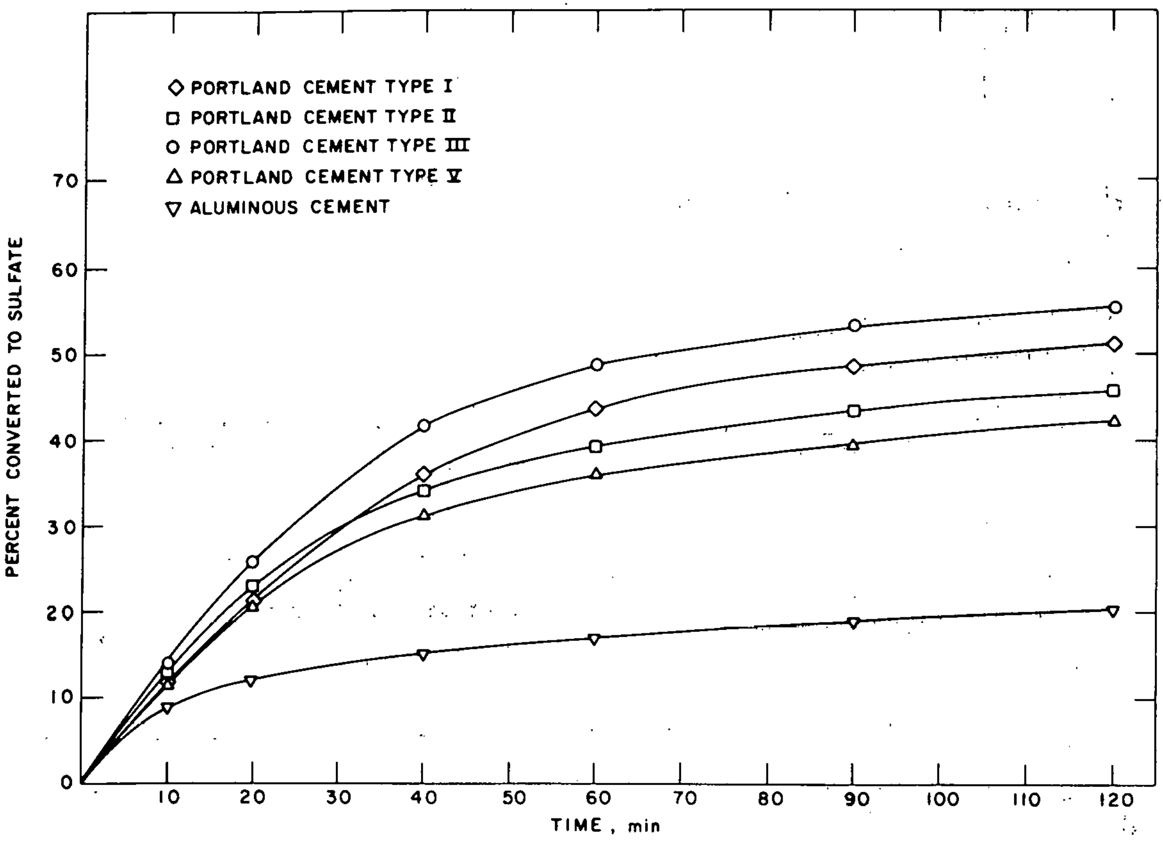

Figure A-1. Sulfation rates of Portland cement and aluminous cement with gas containing 0.25% SO_2, 5% O_2, and 15% CO_2 in N_2 at 900°C.

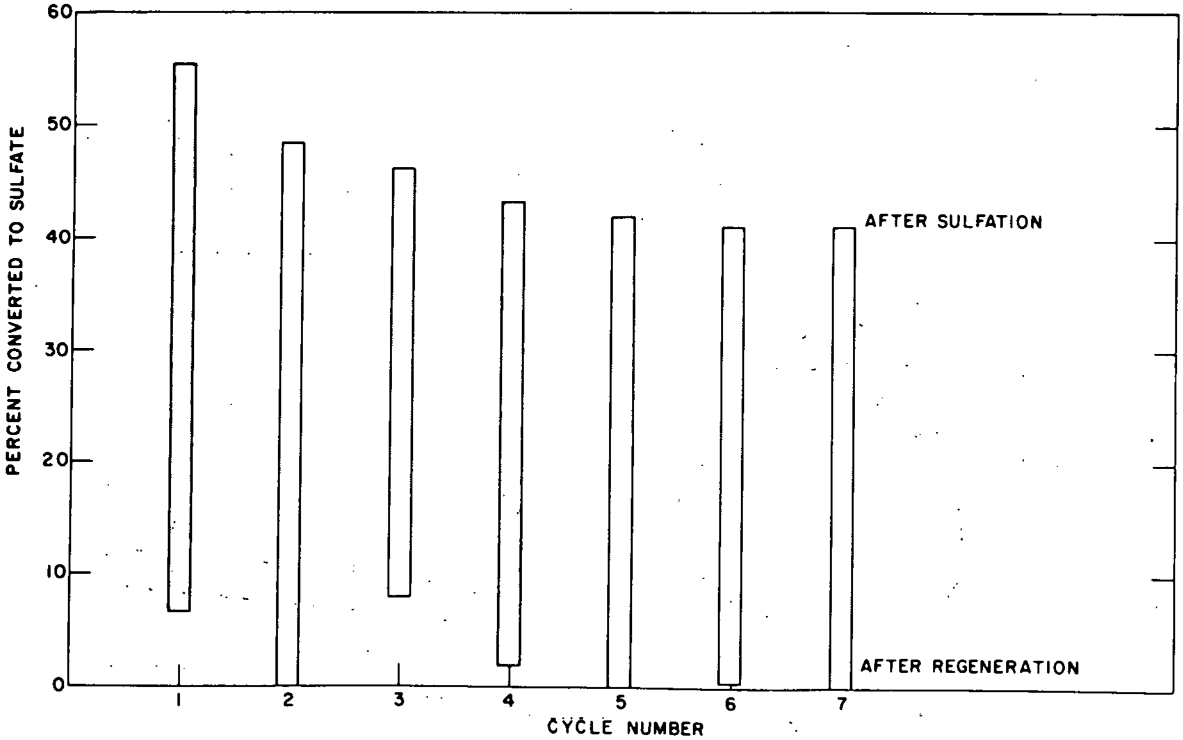

Figure A-2. Cycling of Portland cement powder (type III) between sulfation (0.25% SO_2, 5% O_2, 15% CO_2 and balance N_2 at 900°C) and regeneration (5% CO, 20% CO_2 and 75% N_2 at 1000°C).

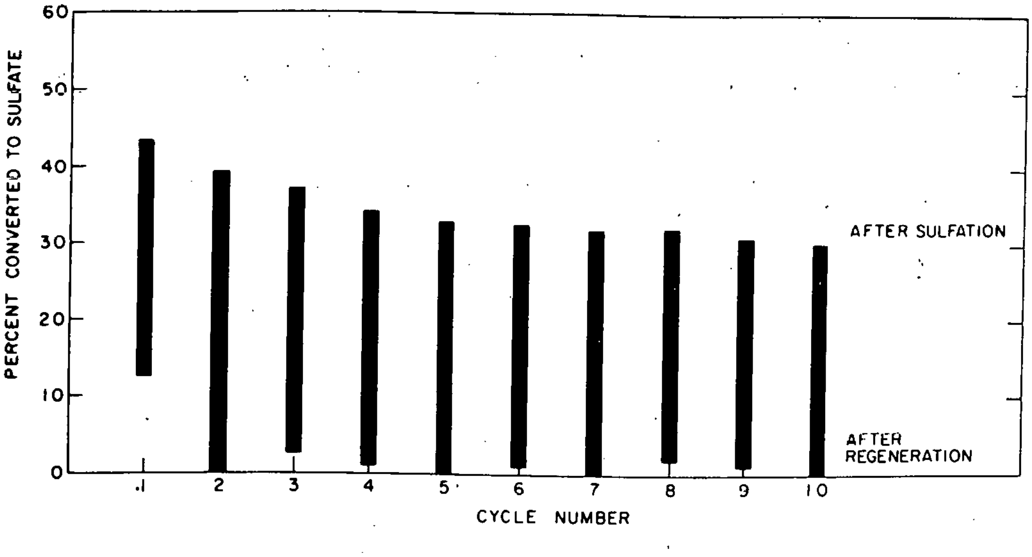

Figure A-3. Cycling of Portland cement extrudates (type III, 16/20 mesh) between sulfation (0.25% SO_2, 5% O_2, 15% CO_2 and balance N_2 at 900°C) and regeneration (5% CO, 20% CO_2 and 75% N_2 at 1000°C).

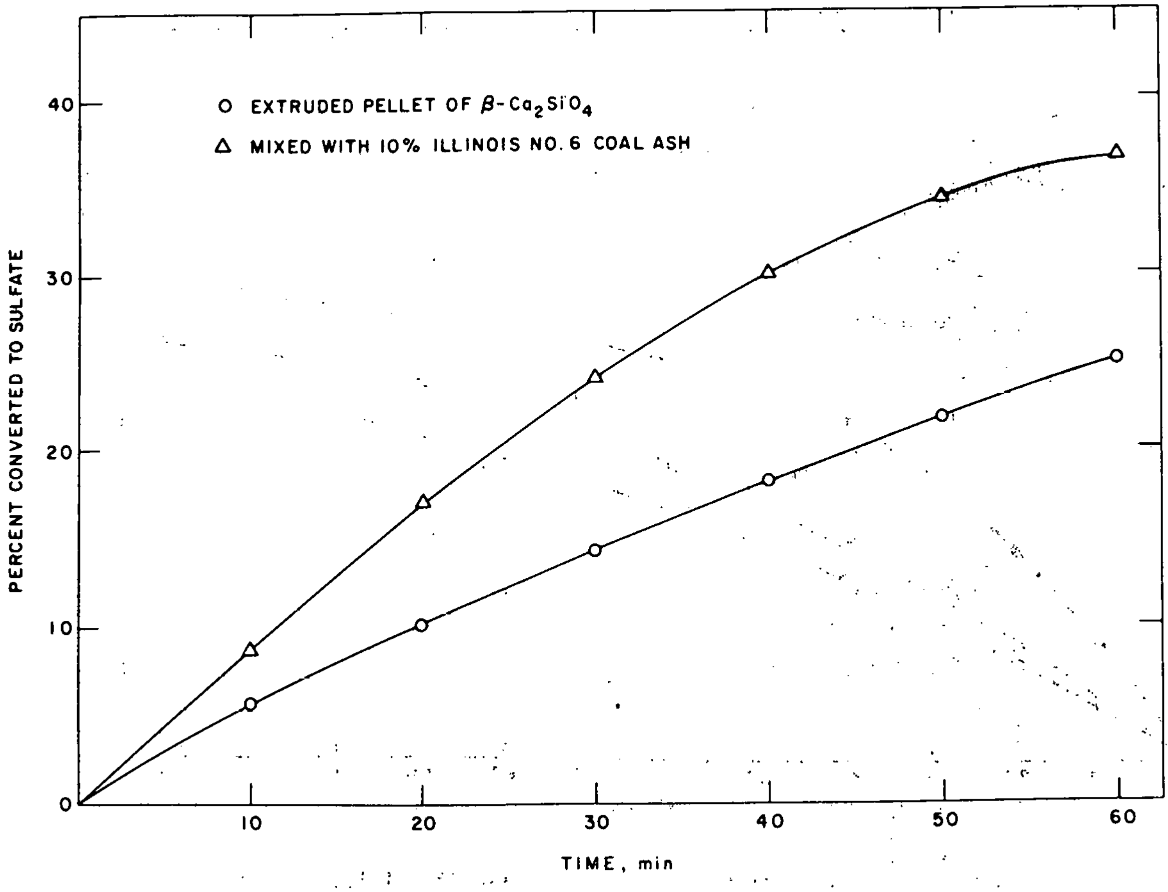

Figure A-4. Sulfation rates of β-Ca_2SiO_4 extrudates (16/20 mesh) with gas containing 0.25% SO_2, 5% O_2 and 15% CO_2 in N_2 at 900°C.

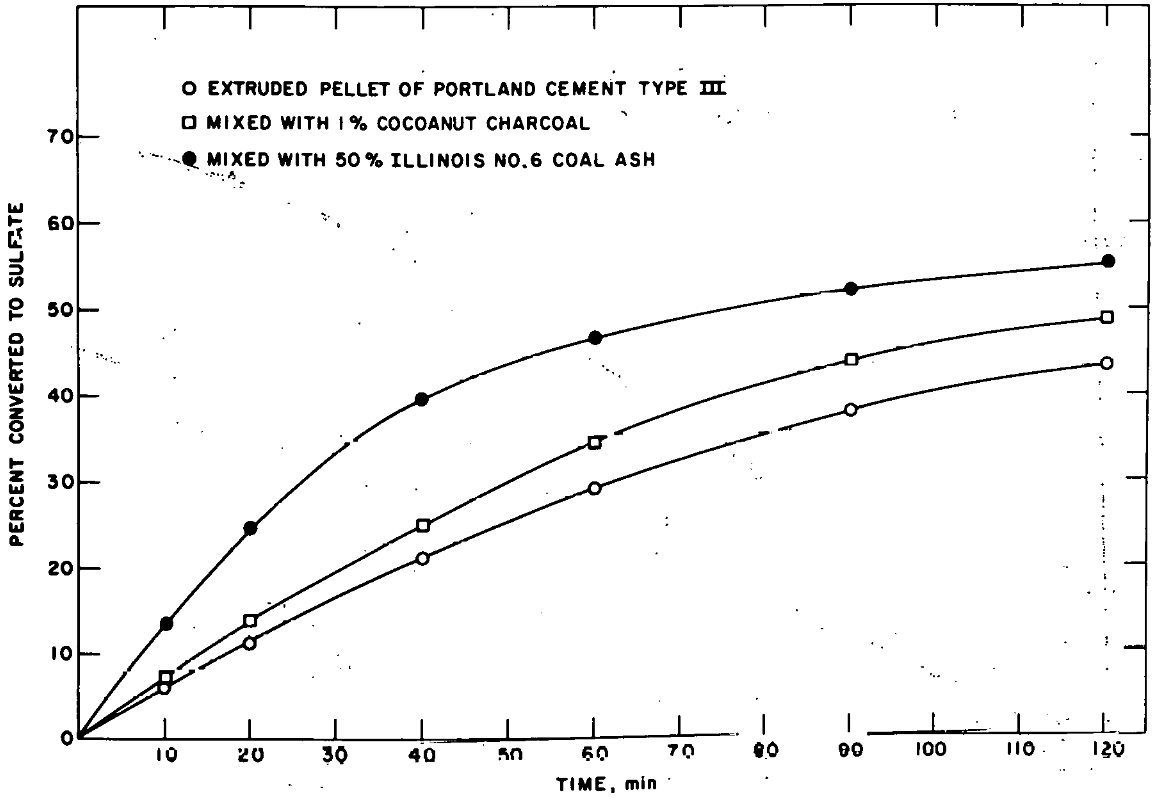

Figure A-5. Sulfation rates of Portland cement type III extrudates (16/20 mesh) with gas containing 0.25% SO_2, 5% O_2 and 15% CO_2 in N_2 at 900°C.

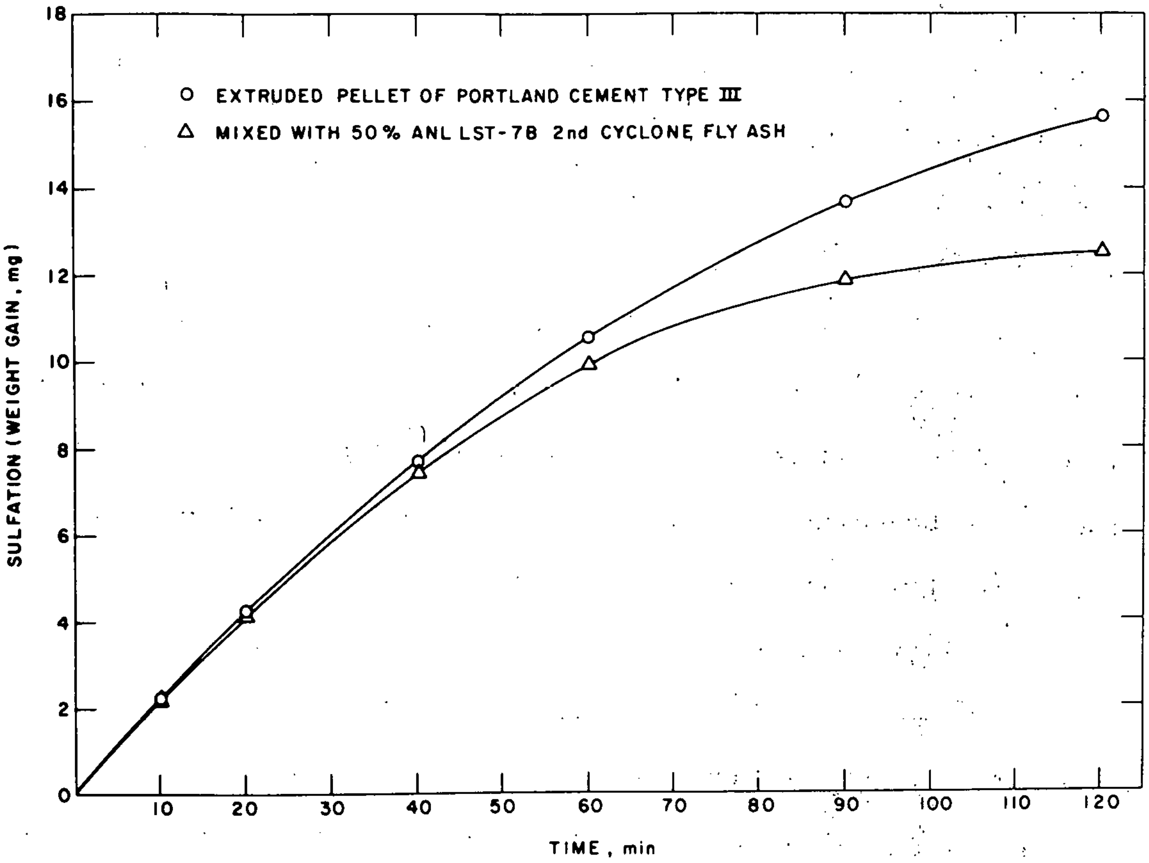

Figure A-6. Sulfation rates of 16/20 mesh extrudates per unit mass of Portland type III and Portland type III with fly ash (1 to 1) at 900°C with 0.25% SO_2, 5% O_2 and 15% CO_2 in N_2.

B. Utilization of Pellets of Limestone Fines for FBC

(J.M. Chen, F.B. Kainz, and G. Farber)

It has been shown that pellets of limestone fine powder are more accessible and reactive to SO_2 than raw stone particles, as shown in Quarterly Progress Report No. 10. However, because the pellets showed little strength to resist attrition in a fluidized-bed, addition of binding materials became necessary in order to enhance the pellet strength. This report summarizes the results of sulfation and attrition tests for pellets of limestone fines with various binders.

Table B-1 summarizes the preparation procedures, composition, and the qualitative strength test of the pellets. In all these preparations, Greer limestone fines with sizes less than 240 U.S. mesh were used. The limestone fines were mixed with binder solutions to form paste. Pellets were formed by extruding the paste samples through a 1/16" ID glass tube. Among these preparations, pellets formed with sodium silicate and colloidal silica showed highest compression strength. However, because these binders, contained sodium ions which may cause corrosion problems, attempts were made to leach the sodium ions by deionized water. It was found that after leaching, pellets formed with sodium silicate lost strength completely, while those formed with colloidal silica retained the original hardness. Thus. among the preparations shown in Table B-1, colloidal silica is the most promising material for effective binding of the limestone fines.

The sulfation experiments were performed in a Cahn TGA system. The apparatus and calculation procedures have been described elsewhere.[3,4] Samples of pellets were calcined in the system under N_2 at 900°C with

25°C/min heating rate. Subsequent sulfation took place with gas composition of 0.25% SO_2, 5% O_2 and balancing N_2 at 900°C and 1 atm pressure. Figure B-1 compares the sulfation rates of the leached limestone/colloidal silica pellets with the raw stone particles. With the same 16/20 mesh particle size, the SO_2 uptake in 2 hour sulfation time was 55% for pellets versus 35% for limestone particles.

The attrition test was performed in a 30 mm fluidized-bed. Both calcined pellets and limestone particles weighing 10 grams with the size of 10/12 mesh were fluidized by 5 ft/sec air velocity at room temperature. The sample weights with sizes greater than 20 mesh were measured as a function of time. Figure B-2 shows the weight loss rate for both samples. The attrition loss for raw limestone particles was found about three times higher than that of pelletized particles.

Cyclic sulfation and regeneration experiments were also performed on pellets of limestone fines with colloidal silica binder in the TGA apparatus. The regeneration condition was 1000°C with 15% CO. Time required for regeneration was generally less than 15 minutes. Figure B-3 shows the extent of CaO conversion to $CaSO_4$ in 6 cycles. No decay in the sulfation activity was observed. After the sixth cycle of regeneration, pellets seems to hold the hardness although no quantitative test were made.

These results clearly indicate that both the strength and the sulfation reactivity are higher for pellets of the limestone fines with colloidal silica than for raw particles.

TABLE B-1

No.	Composition	Cure Method	Comments
1	90% Greer Limestone 350 Mesh Sodium Silicate Base Cement	1/2 Hr at 350°	Strong After Cure, But Weak After Water Washing. Sauereisen #1
2	50% Greer 350 Mesh 50% Sodium Silicate	1/2 Hr at 350°	Same as No. 1. Water Was Used to Make the Paste.
3	55.5% Greer 350 Mesh 16.6 Water 27.7 Phosphoric Acid	1/2 Hr at 300° 3 Hrs. at 950°	Too Weak To Use Crumbles Between Fingers.
4	66% Greer 20% Water 13.3 Sodium Metasilicate	1/2 Hr Air Dry 3 Hrs 950°	Very Weak After Calcining. Strong Before.
6	Greer Mixed Into Paste with Liquid Binder Used in No. 31 Sauereisen Binder is Sodium Silicate and Water	1/2 Hr. Air Dry 950° for 2 Hrs.	Not Very Strong After Calcining.
7	Calcium Aluminate Mix 50% Water + Greer 25% Water + Greer 20% Water + Greer	Air Cure and Heated to 950° for 3 Hrs.	Too Weak to Use. Sauereisen #75
8	75% Greer Limestone 25% Coloidal Silica	Air Dry & Heated to 950° 3 Hrs 240-400 Mesh	Strong After Calcining Ludox Consists of 40% SiO_2 43% Sodium Hydroxide 17% Water.
9	Same as No. 8	2 Hrs at 350°	Washed with Deionized Water Water for 17 Hrs. to Remove Sodium Hydroxide Remains Strong.
10	Same as No. 8	Same	Same as Above, and Analyzed for Sodium 0.189%.
11	Same as No. 8	Same	Not Washed - Analyzed for Sodium - 0.463%.
12	Same	Same	Leached with Deionized Water for 24 Hrs - Recycled 6 Times (James Has Data)
13 A 13 B	Ludox + Greer Limestone Greer Limestone	350° 1/2 Hr	Abrasion Tests Are Being Repeated as 16A, 16B to Have Sharp Edges Removed Before Test.
14	Ludox + Greer	350° 1/2 Hr	Machined to 1/4"x1/2" Cylinder for Compression Test Will Be Compared with Same Dimensions Greer Cylinder.
16 A	Ludox + Greer 240-400 Mesh Extruded and Broken to Pass 12 Held on 20 Mesh	350° 1/2 Hr Calcined at 1000° 1 Hr in Fluid Bed	Rotated at 72 RPM in 4" Dia. Drum Lined with 180 Grit Abrasive Paper to Round Off Edges-Fluidized in 30 mm Bed, Air Flow 20 Liters/min Time Hrs — Wt. Gms. 0 — 9.5356 3 — 9.4760 24 — 9.0277 32 — 8.9217 47 — 8.7665

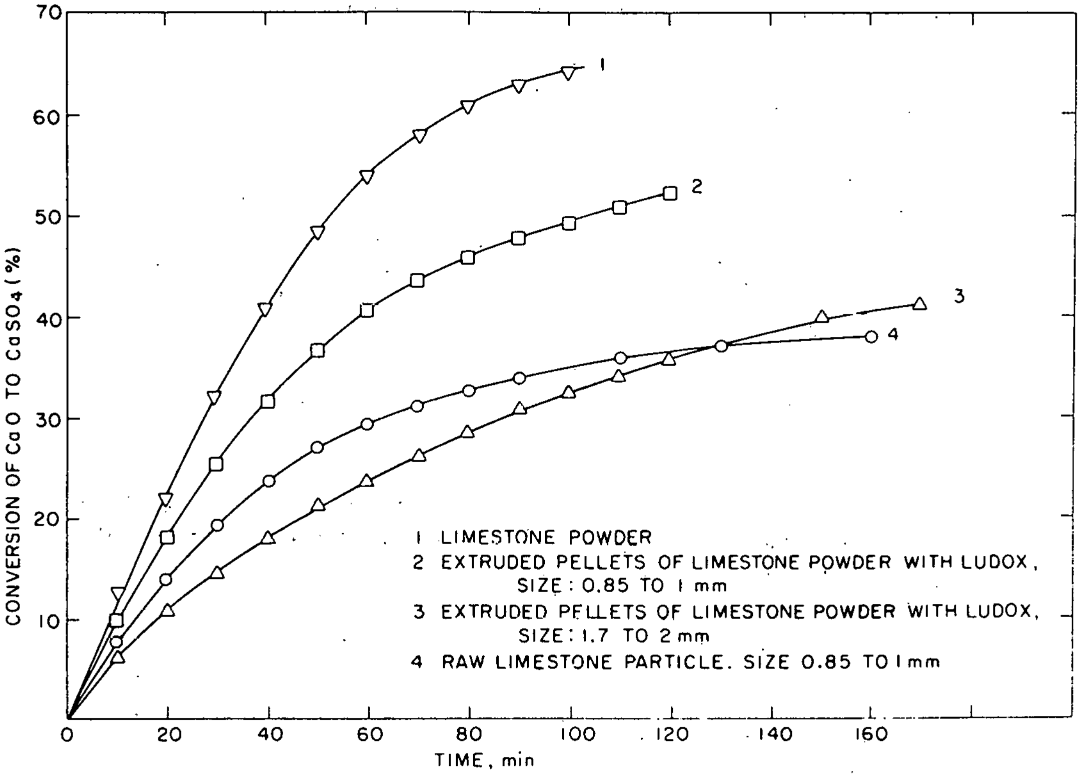

Figure B-1. Comparison of sulfation rates at 900°C in 0.25% SO_2, 5% O_2 and balance N_2.

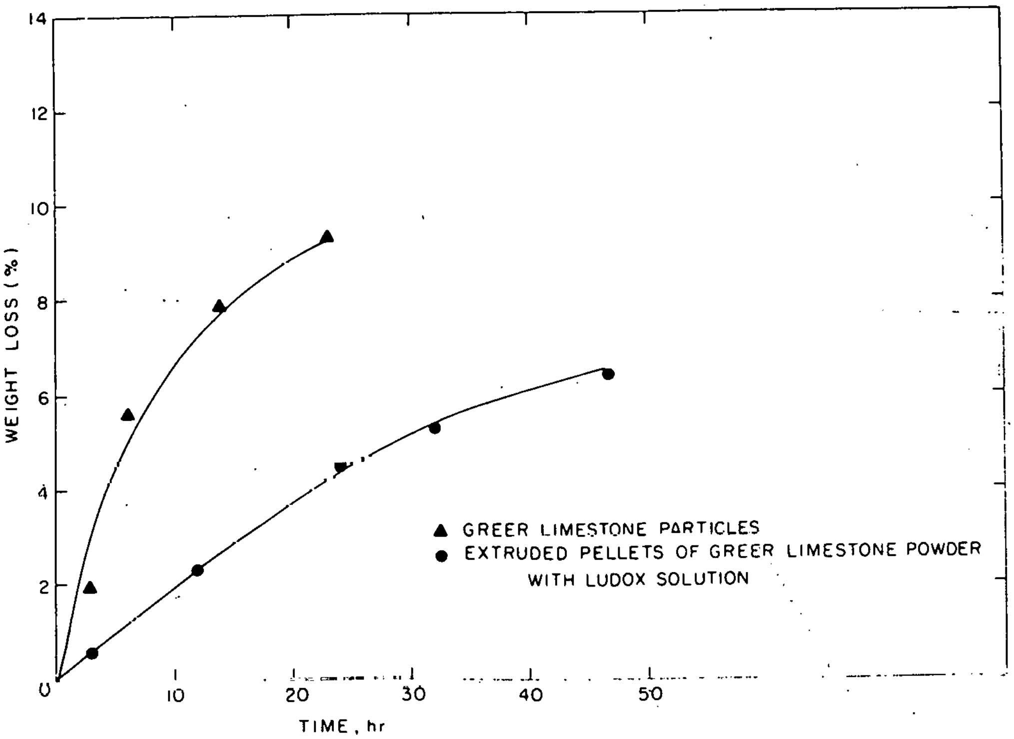

Figure B-2. The attrition weight loss % of 10 gram samples (10/12 mesh) versus time for sample weight losses > 20 mesh.

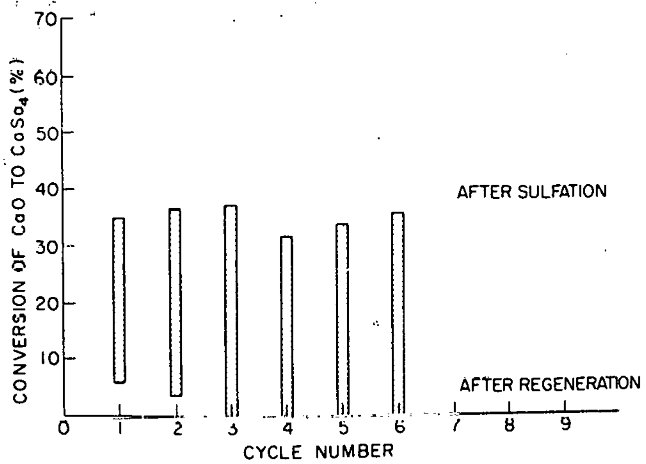

Figure B-3. Cycling of extruded pellets of limestone fines between sulfation (0.25% SO_2, 5% O_2 and balance N_2 at 900°C) and regeneration (15% CO and 85% N_2 at 1000°C).

C. The Temperature of Peaking of Sulfur Retention

(M. Shen, J. Pruzansky, and J.M. Chen)

In a fluidized-bed combustor (FBC), an optimal temperature for maximum sulfur retention has been observed. Generally, these temperatures lie between 830°C and 900°C. Increasing or decreasing the combustion temperatures resulted in lower sulfur retentions. Various explanations for such phenomenon have been proposed. Among these are the existence of the oxidation and reduction zones in the bed,[5] disproportionation reaction of $CaSO_3$,[6] etc.

It has been shown[7] that the pore structure of calcined limestone is strongly affected by the calcination conditions, i.e., the environment of CO_2 partial pressure. The higher the CO_2 partial pressure, the larger the pore size distribution. The reason for this is the CO_2 partial pressure retards the calcination rate, allowing the larger pores to develop. In a fluidized-bed combustor, the CO_2 partial pressure generated at atmospheric pressure is about 0.15 atm. This value relative to the equilibrium CO_2 partial pressure over $CaCO_3/CaO$ is higher at lower temperature. Hence, the retardation effect on calcination is higher at lower temperatures. Thus when calcination occurs at higher temperatures, the lime sorbent will have a lower capacity for SO_2. Combining this effect with the fact that increasing temperature increases the reaction rates, a temperature maximum could conceivably occur for a given solid resisdence time in the bed.

Two sets of experiments were designed to test the above theory. In the first set, limestone samples were calcined and sulfated at the same temperature in the simulated fluidized-bed condition. In the second set of experiments, limestone samples were all calcined at 1000°C, thereby producing the same pore size distribution. Sulfation temperatures were then varied from 750°C to 1000°C.

A DuPont thermo-analyzer Model 951 was used for the sulfation rate measurements. A small quartz boat with an area of about 0.6 cm^2 was used as the sample holder. A quartz tube packed with alumina chips and housed in a clamshell furnace served as the preheater for the reactant gases. About 60 mg of uncalcined Greer limestone (16/20 Tyler mesh) was spread in a thin layer in the quartz boat. The limestone was calcined isothermally in 15% CO_2 and 85% N_2. It was then sulfated isothermally with a simulated combustion gas (0.25% SO_2, 5% O_2 and balance N_2) flowing at a velocity of about 10 cm/sec over the sample surface. This velocity was predetermined sufficiently high to minimize gas film diffusion rates.

Figure C-1 shows the overall rate of the sulfation reaction for limestone calcined at the sulfation temperatures. The percent of sulfation after 2 hours in Figure C-1, was plotted against temperature in Figure C-2. A peaking temperature 875°C is observed. This value is very close to the observed one obtained from FBC tests.

Figure C-3 gives the sulfation rates for Greer limestone calcined at 1000°C. Note that after about 20% sulfation the rates dropped sharply regardless of the sulfation temperatures. Plotting the extent of sulfation after two hours versus temperature as shown in Figure C-4, the sulfation capacity levels off at above 850°C. No peaking temperature is observed. It is reasoned that in about two hours the sulfation was sufficient to plug the transport pores due to the larger molar volume of $CaSO_4$ to CaO, thereby showing no further noticeable increase in reaction rates.

By comparing Figure C-2 and C-4, it is clear that the temperature maximum observed in a FBC can very possibly be due to pore size variations because of calcinations at different temperatures. To further confirm the postulated mechanism, tests of precalcined limestone samples in a fluidized bed are necessary.

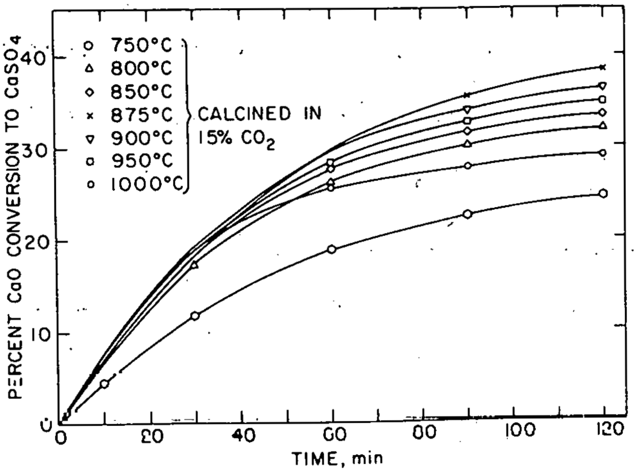

Figure C-1. The effect of temperature on limestone sulfation rate with calcination at the sulfation temperature.

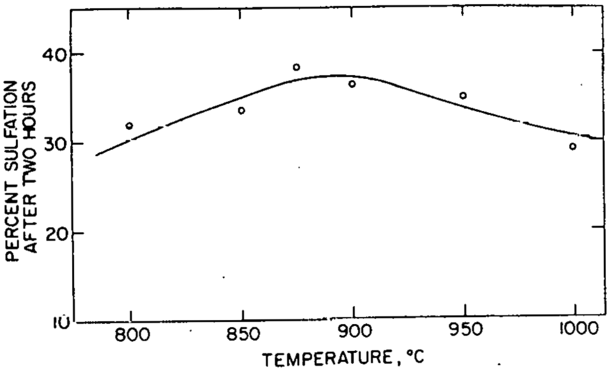

Figure C-2. The effect of temperature on calcium utilization in sulfation with calcination at the sulfation temperature.

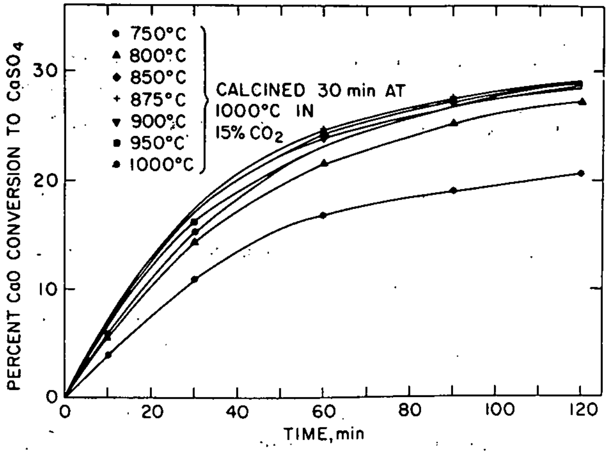

Figure C-3. The effect of temperature on limestone sulfation rate with 30 min. calcination at 1000°C.

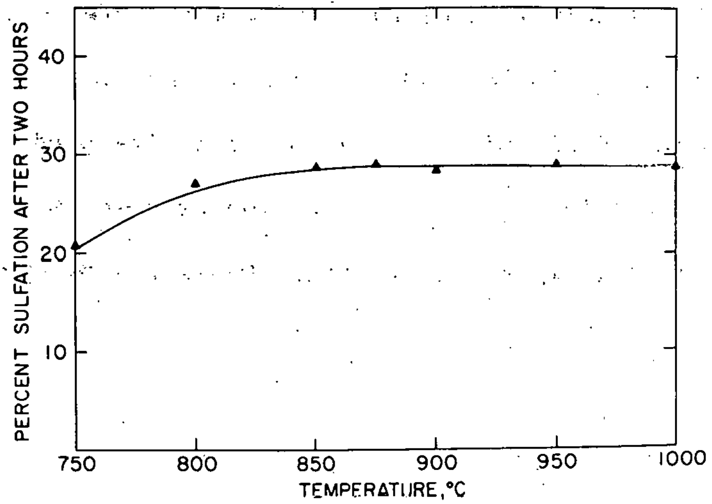

Figure C-4. The effect of temperature on calcium utilization in sulfation with 30 min. calcination at 1000°C.

D. Preparation of Reactive Calcium Silicates

(M. Shen, G. Farber, and J. Pruzansky)

In the previously known technology for the preparation of β-dicalcium silicate, e.g., evaporative decomposition of solutions of calcium nitrate and colloidal silica at 1050°C,[8] or repetitive firing of a mixture of fine powders of calcium carbonate and silica at 1080°C,[9] the temperature is high and therefore, the reactivity is rather low.

A low-temperature solid state process of forming fine particles of reactive β-dicalcium silicate is being investigated by firing a mixture of calcium sulfate (anhydrite or gypsum) and silica together with calcium sulfide (a reducing additive). The temperature of the process described here can be as low as 850°C. This is expected to result in substantial energy savings and to yield more reactive products. Calcium sulfide can be formed by the reduction of calcium sulfate with C, CO, CH_4, H_2 etc. High concentration of SO_2 is the by-product of the process.

E. The Kiln Regeneration Process

A rotary kiln, 3 inch ID, is being constructed with the features of external heating and continuous solid feeding. Fabrication of the rotary kiln will be completed during the next report period. It will be in operation for gathering kinetic and process development data. The regeneration of limestones, calcium silicates and Portland cements using fly ash from FBC will be tested with this device.

Acknowledgement

Comments and guidance provided by Mr. William Fedarko of U.S. Department of Energy are appreciated. Samples of Portland cement clinkers were kindly supplied by Portland Cement Association, Illinois. The colloidal silica "Ludox" was offered by E.I. duPont de Nemours & Co.

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