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## Removal of Sulfur Oxides from Synthetic Flue Gas by Lignite Fly Ash

Ellen Bernard Lukes

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REMOVAL OF SULFUR OXIDES FROM SYNTHETIC  
FLUE GAS BY LIGNITE FLY ASH

by

Mrs. Ellen Bernard Lukes

B. S. in Chemical Engineering, University of North Dakota 1967

A Thesis

Submitted to the Faculty

of the

University of North Dakota

in partial fulfillment of the requirements

for the Degree of

Master of Science

Grand Forks, North Dakota

January  
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This thesis submitted by Mrs. Ellen Bernard Lukes in partial fulfillment of the requirements for the Degree of Master of Science from the University of North Dakota is hereby approved by the Faculty Advisory Committee under whom the work has been done.

/s/ Wayne R. Kube  
(Chairman)

/s/ Robert C. Ellman

/s/ Edward Nelson

/s/ James L. Elder

/s/ A. W. Johnson  
Dean of the Graduate School

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ABSTRACT

Removal of Sulfur Oxides from Synthetic  
Flue Gas by Lignite Fly Ash

by

Mrs. Ellen Bernard Lukes, Master of Science

The University of North Dakota, 1969

Faculty Adviser: Professor Kube

The absorption of sulfur dioxide from a synthetic flue gas by fly ash was investigated. Tests were made by passing a flue gas mixture of known composition over various samples of lignite fly ash and then analyzing the samples by means of x-ray fluorescence.

The significance of time, temperature, partial pressure of sulfur dioxide in the flue gas, and the composition of the ash was determined by statistical analysis. Analysis of variance indicated that the four variables studied all had a significant effect on the amount of sulfur dioxide absorbed by the fly ash at the 99.5 percent confidence level. Five of the six two-way interactions and two of the four three-way interactions were also significant.

At 1,600° F there was approximately a 20 percent increase in the sulfur content (as SO<sub>3</sub>) of certain fly ash samples investigated after 17-1/2 hours of exposure time. Other samples of different initial composition absorbed less SO<sub>2</sub>. This investigation did indicate that a high initial sodium or calcium content increases the amount of SO<sub>2</sub> that fly ash will absorb.

Comparison tests were made using alkalized alumina at 625° F and 1,100° F. Alkalized alumina absorbed sulfur dioxide more readily than fly ash at 625° F. Fly ash was a better absorbent during the first 10 hours of exposure at 1,100° F. At 1,600° F the fly ash absorbed SO<sub>2</sub> as well as the alkalized alumina did at 625° F. Whether the alkalized alumina was in pellet form or crushed, the amount of SO<sub>2</sub> absorbed remained approximately the same over the time intervals studied.

The high temperatures necessary for appreciable absorption of SO<sub>2</sub> by lignite fly ash are such that use in equipment auxiliary to a boiler would not be feasible, but reinjection directly into the boiler may be feasible if the additional ash load can be tolerated.

## INTRODUCTION

Sulfur oxides in flue gas from combustion of fossil fuels present air pollution problems. In addition, absorption by ash particles during combustion may contribute to the formation of fireside deposits in coal fired boilers. Analyses of lignite ash prepared by laboratory ashing procedure indicate that a high percentage of sulfur in lignite is retained in the ash. Limited data from sulfur balances on combustion tests using lignite showed that a lesser percentage is retained during combustion in boilers, and that the retention is related to the composition of the ash.

The primary objective of this research was to investigate the properties of various lignite-derived materials, particularly flyash, in relation to their ability to absorb and retain sulfur oxide containing gases. Sulfur oxide absorption was studied as influenced by the variables of temperature, time, partial pressure of sulfur oxide, and the ash composition. Sulfur content, as  $SO_3$ , in the flyash was determined before and after exposure to a synthetic flue gas.

Tests were run on five flyash materials from lignite from three mines as burned at three different powerplants. These materials represented high- and low-sodium contents in the range of 0.1 to 7.3 percent of ash at levels of calcium concentration between 16.7 and 38.3 percent of ash.

If lignite flyash proved to be a satisfactory absorber of sulfur oxides from flue gases, a use for a waste material would be found and a potentially inexpensive  $SO_2$  removal process might be developed.

## CHAPTER I

### AIR POLLUTION IN THE COAL INDUSTRY

#### Effects of Air Pollution

Air pollution in the United States is costing an estimated \$11 billion annually (\$60 per person) in damage to buildings, equipment, fabrics, crops, livestock and other goods, while also harming the health of the population. In 1963, sulfur dioxide emissions amounted to more than 23 million tons [1]<sup>1</sup>.

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<sup>1</sup>Numbers in brackets refer to items in List of References.

Other physical and chemical methods for removing pyrite sulfur from coal, which are in experimental stages and seem promising, are [3]:

- a. stage pulverization and subsequent removal of pyrites (Bituminous Coal Research, Inc.)
- b. electrostatic separation of pyrites (Battelle Memorial Institute)
- c. froth floatation using addition of depressants specific to pyrites (Bureau of Mines)
- d. magnetic separation of pyrites (Coal Research Bureau, West Virginia University, and Bureau of Mines)
- e. fluidized bed with air elutriation (Bureau of Mines)

3. Use of dolomite and other additives to the furnace [3]: The results of injecting finely divided material into existing boiler systems or blending limestone with the fuel have generally been discouraging in terms of the ability to obtain high efficiency and high utilization of the material added. Conflicting reports have circulated as to the effectiveness of limestone addition. These reports range from complete to negligible removal of sulfur oxides.

4. High stacks [3]: Tall stacks can provide relief in localized situations, but they are appropriate only for large installations where their cost can be justified. Chimney heights of 800 feet are becoming quite common. The objective is to keep ground level concentrations sufficiently low to avoid any damageable or harmful effects. This solution does not solve the pollution problem, but can give local relief.

5. Gas treating processes (wet and dry) [5]: As sulfur oxides are acidic, alkaline media are used in most of the schemes for their removal. Most of the work on wet processes has been discontinued because of metal corrosion, cooling of the stack gases, loss of buoyancy, and high costs of operation and waste disposal.

Chemical processes can achieve a high degree of sulfur removal, but as yet no method appears to have overcome the problems of loss of part of the coal, loss of energy in the coal, or the high costs of treatment.

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elemental sulfur by the conventional Claus process. The principle difficulty in the process is the loss of alkalized alumina by attrition over extended periods of time [4].

It is felt that the Bureau's process has a distinct advantage in that elemental sulfur is the end product. Elemental sulfur does not present any special handling problems, as do the acid products realized from other processes.

## CHAPTER II

### DESIGN OF APPARATUS

The overall functions required for the present experiments were to pass a synthetic flue gas containing sulfur dioxide over different samples of fly ash. The amount of  $\text{SO}_2$  absorbed was determined for various time intervals by analyzing for the increase in  $\text{SO}_3$  content in the ash. A schematic diagram showing the test arrangement is given in figure 1.

#### Reaction Vessel

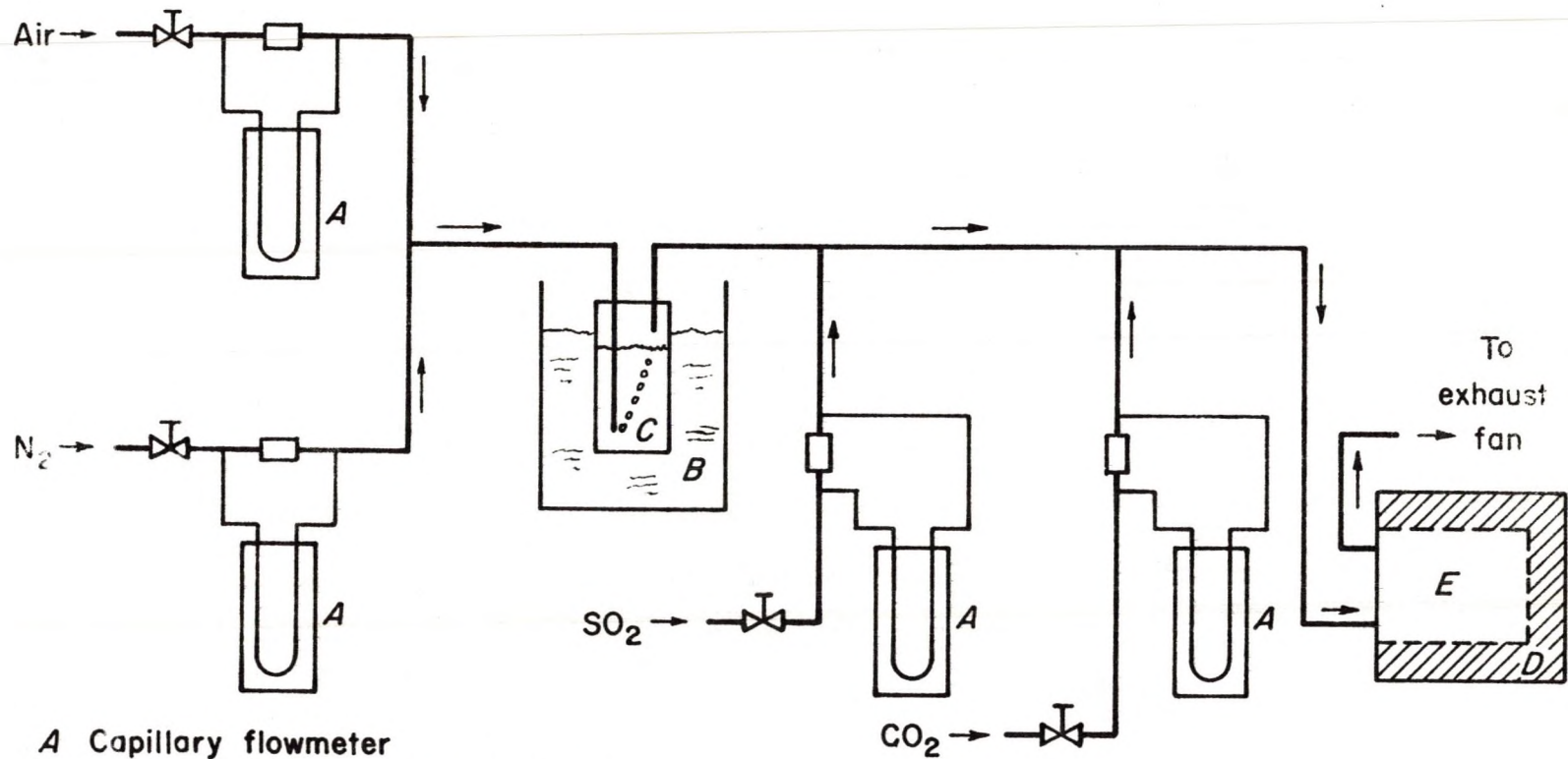
An airtight stainless steel box with two shelves was used to contain the fly ash samples as a synthetic flue gas was passed over them. The design and dimensions are given in figure 2.

The flue gas enters through perforated stainless steel tubing along the upper right length of the box. Previously the gas has been passed through tubing on the right outside wall in order to preheat the synthetic flue gas before it passes through the box.

The flue gas leaves through another perforated tube along the bottom left length of the box. The tubing then leads to the exhaust fan.

#### Flowmeters

The gases entering the box were measured using capillary flowmeters. The difference in pressure on the two sides of a capillary tube (as indicated by a manometer) is a measure of the rate of flow of gas through the flowmeter. The flowmeters were calibrated at several different rates of flow, and a smooth curve was drawn showing the rate of flow as a function of the difference in levels. The calibration curves can be found in Appendix C, figures 17 through 20.



- A** Capillary flowmeter
- B** Constant temperature water bath
- C** Distilled water
- D** Burrell electric furnace
- E** Flyash sample container, see Fig

Fig 1 Schematic design of apparatus and control system for laboratory study of SO<sub>2</sub> absorption by flyash.

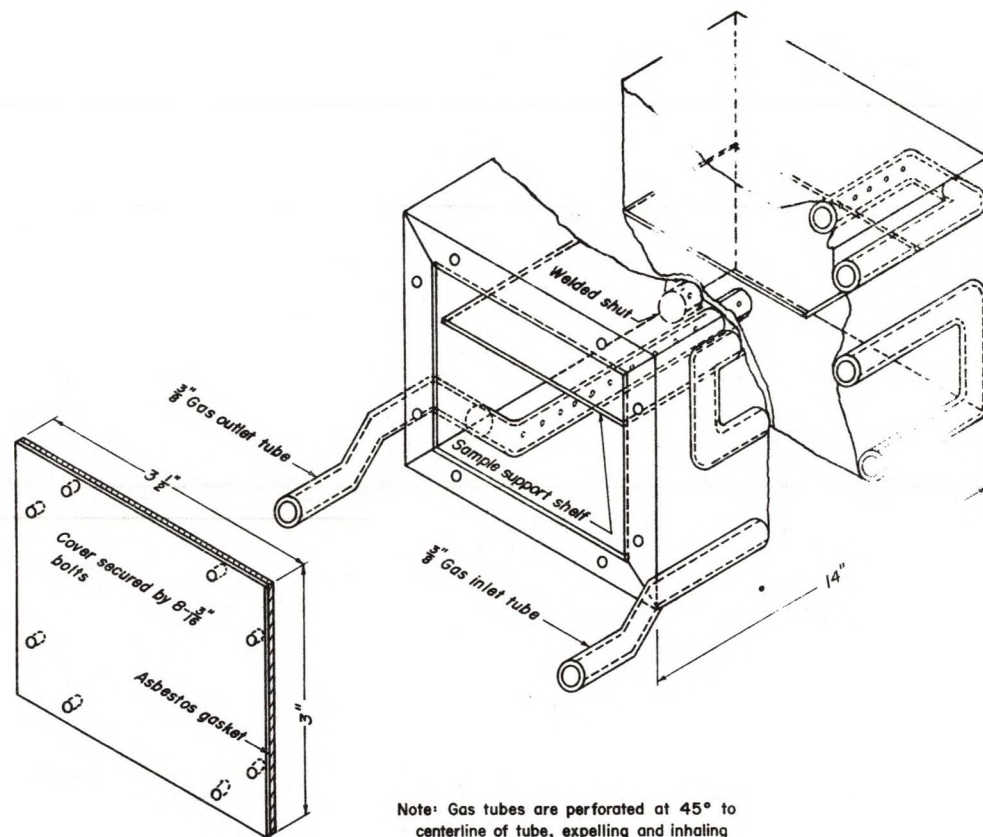


Fig 2 Flyash sample container.

In calibrating the SO<sub>2</sub> capillary, the quantity of SO<sub>2</sub> for a known time interval was measured by bubbling the gas through a 3 percent solution of hydrogen peroxide. The quantity of SO<sub>2</sub> present in the solution was then calculated by titrating with a standard base (0.1 N NaOH) using methyl purple as the indicator. The calibration curve was drawn from this data (figure 20).

#### Moisture Control

Moisture was added to the atmosphere by bubbling the air and nitrogen through a thermostated flask of distilled water. A constant temperature bath around the flask of distilled water controlled the vapor pressure of the water.

#### Electric Furnace

The entire box was placed in a Burrell Electric Furnace (figure 3). The high temperature electric furnace was manufactured by the Burrell Corporation, Pittsburgh, Pennsylvania. Temperature in the furnace was automatically controlled by a millivoltmeter coupled to a platinum-platinum 13 percent rhodium thermocouple. The heating elements consisted of eight silicon carbide rods spaced so as to give a uniform temperature profile throughout the usable furnace area [6].

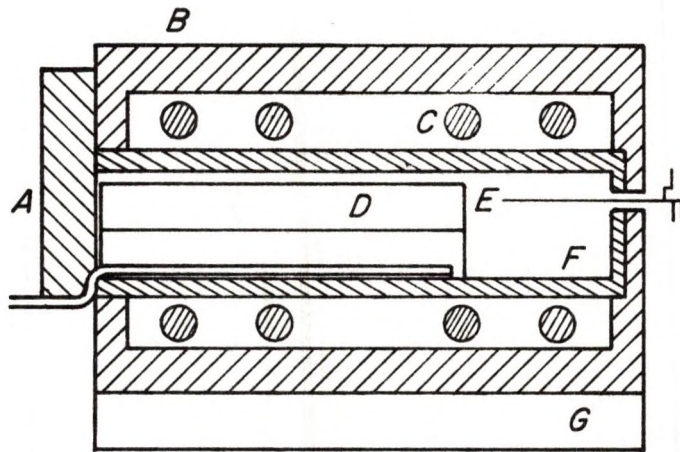
### CHAPTER III

#### EXPERIMENTAL PROCEDURE

The absorption of sulfur dioxide by lignite fly ash exposed to synthetic flue gas at a constant temperature was measured by analyzing the ash for SO<sub>3</sub> content prior to and after exposure. The amount of absorption in relation to time was determined by exposing similar samples for different periods. Variables studied were time, temperature, partial pressure of sulfur dioxide, and the composition of the fly ash.

The synthetic flue gas consisted of: nitrogen 43 percent, air 33 percent, carbon dioxide 12 percent, water vapor 12 percent, and sulfur dioxide of 600, 1,200, or 1,800 parts per million. Since the concentration of sulfur dioxide was always only a fraction of 1 percent, changes in its concentration had no measurable effect on the remaining components, which were maintained at the foregoing percentages.

The lengths of time over which different samples of similar ash were exposed were 1/2, 1-1/2, 3-1/2, 7-1/2, and 17-1/2 hours.



- A* Door
- B* High temperature insulating firebrick
- C* Silicon carbide heating element
- D* Flyash sample container, see Fig
- E* Pt/Pt 13 percent Rh thermocouples
- F* Cast high temperature hearth plate
- G* Support for furnace

Fig 3 Burrell high temperature electric furnace.



The temperature levels selected were 350°, 800°, 1,100°, and 1,600° F. A temperature of 350° F corresponds to the temperature of the combustion gases leaving the air-heater in a boiler, and 800° F to the temperature entering the air heater. A temperature of 1,100° F has been reported as the point of maximum absorption of sulfur dioxide by bituminous coal ash [7], and 1,600° F corresponds to a temperature beyond this reported maximum absorption temperature. An additional test was run at 625° F as a comparison with the Bureau of Mines process using alkalized alumina.

The major test series were run on five fly ash materials relating to lignite from three mines, and burned at three different powerplants. These materials represented high- and low-sodium contents in the range of 0.1 to 7.3 percent of the ash at levels of calcium content between 16.7 and 38.3 percent of the ash. The fly ash was sampled and used as recovered from the fly ash collectors.

Each 4-gram sample of fly ash was placed in a porcelain boat to a depth of approximately 1/4 inch. For each run twenty-five boats were placed on two shelves in the fly ash sample container (Figure 2) and inserted into the cold furnace. After heating to the required temperature, the flow of flue gas was started at 0.39 cubic feet per minute at 80° F. After specific time intervals fly ash samples were withdrawn and placed in a desiccator to cool.

The fly ash samples, after cooling, were analyzed for SO<sub>3</sub> by x-ray fluorescence using a Philips vacuum x-ray spectrograph equipped with a chromium x-ray tube and a flow proportional counter.

In order to find if any other component of the fly ash other than SO<sub>3</sub> changed appreciably, a complete analysis of the fly ash used was made before and after an experimental run. No indications of change in other constituents were noted. These data can be found in Appendix B, Tables 7 through 18.

Tests were designed with a high flow rate for the purpose of supplying a large excess of sulfur dioxide over that which could be absorbed. In this manner the concentration of sulfur dioxide in the container was maintained at a level close to that in the entering stream. In a test at 1,100° F and 1,200 parts per million sulfur dioxide, representing one of the higher rates of absorption, analysis of the exit gas indicated that 70 percent of the sulfur dioxide admitted remained in the exit stream.

Duplicate tests were run to determine whether the position of a fly ash sample within the container had an important effect on the amount of sulfur dioxide absorbed. Table 1 shows absorption in two tests for fly ash sample No. 4 at 1,100° F and 1,200 parts per million sulfur dioxide. The difference between A, the usual sample position, and B,

The details of the method of factorial analysis are outlined in Table 2. The sum of squares listed opposite each of the test variables (designated factors S, A, F, and T) represent values for the main effects of the independent variables. Similarly, sums of squares given for second and third order interactions represent effects of varying levels of independent variables on each other. For example, the interaction S x T indicates an influence of changing levels of SO<sub>2</sub> partial pressure and of furnace temperature not accounted for by the sum of squares for S and for T. The residual sum of squares relates to the uncontrolled variation or error, and since no replicates were performed at the various levels, this term takes the place of a fourth order interaction.

From the sums of squares in Table 2, the mean squares or variances are obtained by dividing by the degrees of freedom. Then by using the "F" test, each of the mean squares for the factors and their interactions are compared with the mean square for the residual (representing error) by calculating a ratio with the residual sum of squares as the denominator to determine their individual significance. This ratio is termed "F" and is compared to critical values which have previously been calculated and tabulated for the desired confidence level and the appropriate degrees of freedom.

## CHAPTER IV

### EXPERIMENTAL RESULTS AND DISCUSSIONS

This chapter describes the results obtained in the investigation of the absorption of sulfur dioxide from a synthetic flue gas by fly ash. A complete set of data is included in the Appendix.

#### Statistical Analysis of the SO<sub>2</sub> Absorption Data

The significance of the variables regarding their effect on the amount of sulfur dioxide absorbed is described by a sum of squares statistical analysis of the data. The statistical approach allows statements regarding the significance of the variables to be made with certain confidence. The analysis of variance for the amount of sulfur dioxide absorbed presented in Table 3 is patterned after Wine [8].

The ratio of the mean square terms of the other sources of variation to the residual mean square is the value of F for that particular source of variation. In a fixed effects experiment, such as this, the computed F value is then compared to the F value which is expected at the degrees of freedom involved and the degree of confidence desired. In order to be significant at this confidence level, the calculated F must be larger than the expected or critical F value.

TABLE 3

ANALYSIS OF VARIANCE OF SO<sub>2</sub> ABSORPTION DATA

Source of variation <sup>a</sup>	Sum of squares	Degrees of freedom	Mean square	Calculated "F"	Critical "F"
S	108.92	2	54.46	188.97 <sup>b</sup>	5.64
A	80.58	4	20.14	69.90 <sup>b</sup>	4.01
F	931.99	3	310.66	1,077.95 <sup>b</sup>	4.59
T	560.74	4	140.18	486.41 <sup>b</sup>	4.01
S x T	47.10	8	5.89	20.43 <sup>b</sup>	3.01
A x T	76.00	16	4.75	16.48 <sup>b</sup>	2.43
F x T	404.15	12	33.68	116.86 <sup>b</sup>	2.62
S x A	4.19	8	0.52	1.82	3.01
A x F	54.52	12	4.54	15.76 <sup>b</sup>	2.62
S x F	190.94	6	31.82	110.42 <sup>b</sup>	3.36
A x F x T	116.48	48	2.43	8.42 <sup>b</sup>	1.83
S x F x T	81.83	24	3.41	11.83 <sup>b</sup>	2.17
S x A x T	6.06	32	0.19	0.66	2.04
S x A x F	10.36	24	0.43	1.50	2.17
Residual	27.66	96	0.29		
Total	2,701.53	299			

<sup>a</sup>S = SO<sub>2</sub> partial pressure      F = furnace temperature  
A = ash composition              T = time intervals

<sup>b</sup>Significant at 99.5 percent confidence level.

The analysis of variance shows the sulfur dioxide partial pressure (S) to have a significant effect on the amount of sulfur dioxide absorbed at a confidence level of 99.5 percent. This indicates, for this data, that a statement saying, "the partial pressure of sulfur dioxide in the synthetic flue gas has a significant effect on the amount of sulfur dioxide absorbed by the flyash samples," would be correct 99.5 times out of 100.

The five different fly ash samples (the A factor) also had a significant effect on the amount of sulfur dioxide absorbed. The variation caused by the fly ash compositions is again significant at the 99.5 percent confidence level.

The third factor investigated, the furnace temperature (F), is also significant at the 99.5 percent confidence level. The computed "F" value of 1,077.95, the largest "F" value computed for any of the factors or interactions involved, indicates that the greatest portion of the variation observed in the amount of sulfur dioxide absorbed by the fly ash samples is due to the temperature factor.

There were five different samples of fly ash used in this research project. Samples No. 1 and 4 appeared to absorb more sulfur dioxide than the other samples tested. Sample No. 4 had the highest calcium content and sample No. 1 had the highest sodium content of the five different fly ash samples tested. It has been thought that the calcium and sodium content of the fly ash influences its sulfur dioxide absorbing tendencies. The present data seemed to support this idea. In order to give conclusive reasons for the higher absorption, more investigation of other samples of fly ash would be needed.

Sample No. 5, which is a low sodium content fly ash, absorbed very little sulfur dioxide until the temperature reached 1,600° F. After 17-1/2 hours and at 600 and 1,200 parts per million sulfur dioxide, sample No. 5 absorbed more SO<sub>2</sub> than sample No. 4. The temperature greatly affects the amount of sulfur dioxide absorbed for the samples of different ash composition.

The high temperatures required for absorption by lignite fly ash would likely discourage its use in absorption equipment auxiliary to a boiler but would still allow absorption by the reinjection of fly ash directly into the boiler if this can be tolerated with reference to boiler fouling. But it must also be kept in mind that the fly ash has an initial sulfur content from prior passage through a boiler in contact with the sulfur dioxide in the flue gas.

Unlike the fly ash tested by Orning [7], a maximum absorption temperature was not reached within the range 350° to 1,600° F, as shown by figure 14, Appendix C. Orning achieved a maximum absorption temperature of 1,100° F for fly ash from bituminous coals. Lignite fly ash continued to absorb sulfur dioxide at an increased rate at 1,600° F in the present tests. Further investigation could be made as to whether an inflection point exists in the absorption versus temperature plot at temperatures exceeding 1,600° F. Another reaction vessel would be necessary as the present unit is limited, as designed, to approximately 1,600° F. In addition, higher service temperature gaskets and stainless steel screws would be required.

#### Comparison Tests Using Alkalinized Alumina

A process for removing sulfur dioxide from stack gases using alkalinized alumina as the absorbent has been studied at the U.S. Bureau of Mines, Pittsburgh, Pa. [9]. Tests were run to compare the absorption rate of fly ash with samples of alkalinized alumina.

The alkalinized alumina is manufactured in pellet form (about 1/16 inch in diameter), and the fly ash is not. A test was made to see if the alkalinized alumina absorption increased if the sample was crushed

occur as a function of the sulfur dioxide concentration in the flue gas, but as the temperature level is increased, the samples become somewhat caked, and a red-brown porous crust forms on the surface. At 1,600° F this reddish layer comprised almost one-fourth of the sample.

Under the crust in sample No. 4 was a powdery substance, almost grey in color. The bottom layer of each fly ash sample which absorbed the most sulfur dioxide [9] consisted of extremely fine particles with a definite color change from the original ash or the top layer. Fly ash samples No. 3 and 5, which were the poorer absorbers of the five samples studied, formed a harder cake than the other fly ash samples, but still remained porous and were easily broken. Porosity of all samples was high and the inner portion was apparently available to the SO<sub>2</sub> in the gas stream.

### CONCLUSIONS

The conclusions that follow are based on the author's interpretation of the results of the investigation:

In order to be used as an absorbent for sulfur dioxide, lignite fly ash would have to be utilized at elevated temperatures. In the temperature interval studied, 350° to 1,600° F, the fly ash absorbed more sulfur dioxide at 1,600° F than at any of the lower temperatures tested. In order to be useful commercially, fly ash could be reinjected directly into the boiler, if this could be tolerated with reference to boiler fouling. An advantage of fly ash is that the absorption rate is highest during initial time intervals.

Unlike the fly ash from bituminous coal studied by Orning [7], no maximum absorption temperature was found between 350° and 1,600° F. The bituminous fly ash studied by Orning exhibited a maximum absorption temperature at 1,100° F. The bituminous fly ash tested either above or below 1,100° F absorbed less sulfur dioxide. Using fly ash from lignite, the amount of sulfur dioxide absorbed continued to rise as the temperature increased to 1,600° F.

The statistical analysis of the sulfur dioxide absorption data showed that each of the four variables tested had a significant effect on the amount of sulfur dioxide absorbed by the fly ash. The longer the time, the higher the temperature, the higher the partial pressure of sulfur dioxide in the flue gas--the more sulfur dioxide the fly ash samples will absorb. The composition of the fly ash also had an effect on the amount of sulfur dioxide absorbed. Tests showed that a high calcium or sodium content in the fly ash increased the absorption of sulfur dioxide. Further tests should be conducted to determine the effect of temperature above 1,600° F and to investigate more thoroughly the influence of ash composition.

## APPENDIX A

### SYMBOLS

- $A_1$  = Fly ash sample No. 1 (Hoot Lake sample).  
 $A_2$  = Fly ash sample No. 2 (Hoot Lake sample).  
 $A_3$  = Fly ash sample No. 3 (Voltaire sample).  
 $A_4$  = Fly ash sample No. 4 (Voltaire sample).  
 $A_5$  = Fly ash sample No. 5 (Sidney sample).  
 $F_1$  = Furnace temperature of 350° F.  
 $F_2$  = Furnace temperature of 800° F.  
 $F_3$  = Furnace temperature 1,100° F.  
 $F_4$  = Furnace temperature 1,600° F.  
 $S_1$  = 600 parts per million sulfur dioxide in flue gas.  
 $S_2$  = 1,200 parts per million sulfur dioxide in flue gas.  
 $S_3$  = 1,800 parts per million sulfur dioxide in flue gas.  
 $T_1$  = Cumulative time of 1/2 hour.  
 $T_2$  = Cumulative time of 1-1/2 hours.  
 $T_3$  = Cumulative time of 3-1/2 hours.  
 $T_4$  = Cumulative time of 7-1/2 hours.  
 $T_5$  = Cumulative time of 17-1/2 hours.  
 $SO_2$  = Sulfur dioxide.  
 $SO_3$  = Sulfur trioxide.  
PAA = Alkalized alumina in pellet form (1/16 inch diameter).  
CAA = Crushed alkalized alumina.

## APPENDIX B

TABLE 5  
PERCENT INCREASE OF SO<sub>3</sub> IN THE ASH

		S <sub>1</sub>					S <sub>2</sub>					S <sub>3</sub>				
		A1	A2	A3	A4	A5	A1	A2	A3	A4	A5	A1	A2	A3	A4	A5
F <sub>1</sub>	T <sub>1</sub>	0.4	0.1	0.3	0.3	-0.1	0.1	0.1	0.2	0.2	-0.1	0.1	0.0	0.2	0.1	-0.3
	T <sub>2</sub>	0.3	0.3	0.4	0.6	-0.1	0.2	0.2	0.4	0.3	-0.2	0.1	0.4	0.2	0.3	-0.3
	T <sub>3</sub>	0.1	0.4	0.5	0.6	0.0	0.1	0.3	0.5	0.4	-0.1	0.1	0.2	0.4	0.2	-0.1
	T <sub>4</sub>	0.2	0.8	0.4	0.5	-0.1	0.1	0.3	0.4	0.6	0.0	0.5	0.4	0.4	0.6	-0.1
	T <sub>5</sub>	0.5	0.7	0.5	0.8	-0.1	0.5	0.4	0.5	0.6	-0.1	0.7	0.6	0.5	0.8	0.0
F <sub>2</sub>	T <sub>1</sub>	0.6	0.5	0.4	0.5	-0.1	0.2	0.6	0.4	0.3	-0.2	0.9	0.3	0.4	0.5	-0.1
	T <sub>2</sub>	0.8	0.6	0.4	1.0	-0.1	1.0	0.6	0.6	1.3	-0.1	0.7	0.6	0.8	0.2	0.0
	T <sub>3</sub>	1.0	0.7	0.7	1.3	0.0	1.2	0.7	0.8	1.7	0.0	1.8	1.0	1.1	1.7	0.0
	T <sub>4</sub>	1.2	0.7	1.0	2.4	0.0	1.8	0.9	1.1	2.4	0.1	2.1	0.9	1.2	2.8	0.2
	T <sub>5</sub>	3.0	1.1	1.3	3.2	0.2	2.9	1.5	1.5	3.4	0.3	3.4	1.8	1.7	3.5	0.2
F <sub>3</sub>	T <sub>1</sub>	1.2	0.8	0.7	0.8	0.2	1.1	1.0	0.7	0.8	0.0	2.0	1.0	0.7	1.9	0.1
	T <sub>2</sub>	1.7	1.0	0.7	1.4	0.2	1.8	1.4	1.1	2.1	0.0	2.5	1.8	1.2	3.3	0.6
	T <sub>3</sub>	2.1	1.7	1.4	2.6	0.5	3.2	2.1	1.7	3.0	0.0	4.1	2.1	1.9	4.2	0.6
	T <sub>4</sub>	4.0	2.4	1.7	4.8	1.5	4.3	2.1	1.9	5.8	1.6	5.7	1.7	2.9	6.6	2.7
	T <sub>5</sub>	6.6	4.4	3.5	6.5	3.1	6.0	5.2	4.2	8.6	4.6	9.6	6.0	4.6	9.1	5.3
F <sub>4</sub>	T <sub>1</sub>	0.0	0.6	0.7	0.2	0.8	1.3	1.6	1.0	1.0	1.1	0.9	1.7	1.4	0.8	1.8
	T <sub>2</sub>	-0.2	0.8	1.3	1.9	1.6	1.8	3.6	2.4	3.1	2.3	2.0	3.6	2.4	4.7	3.1
	T <sub>3</sub>	0.9	2.6	2.6	2.4	2.8	3.3	4.9	4.9	5.4	4.6	2.5	7.7	6.3	6.7	9.0
	T <sub>4</sub>	4.0	4.1	1.9	3.3	3.0	9.0	10.2	5.8	7.8	6.5	12.5	12.4	7.5	11.4	8.5
	T <sub>5</sub>	6.9	2.4	2.4	4.1	5.0	17.4	8.5	7.1	9.1	10.8	19.4	10.8	9.3	14.5	12.2

TABLE 8

ANALYSIS OF SAMPLES (IN PERCENT) AFTER 17-1/2 HOURS  
AT 350° F AND 1,200 PARTS PER MILLION SO<sub>2</sub>

Component	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	A <sub>5</sub>
Fe <sub>2</sub> O <sub>3</sub>	11.50	12.90	6.90	6.50	7.65
TiO <sub>2</sub>	0.83	0.72	0.90	0.69	0.81
CaO	25.70	29.90	17.20	36.20	27.65
K <sub>2</sub> O	0.51	0.36	0.74	0.41	0.82
SO <sub>3</sub>	6.28	4.50	1.60	4.92	1.73
P <sub>2</sub> O <sub>5</sub>	0.80	0.91	0.52	1.12	0.87
SiO <sub>2</sub>	26.40	27.27	45.45	24.40	34.10
Al <sub>2</sub> O <sub>3</sub>	13.85	15.0	16.85	11.20	18.25
MgO	7.85	8.15	4.95	7.60	9.80
Na <sub>2</sub> O	5.50	2.00	5.40	4.60	0.20
Total	99.22	101.71	100.51	97.64	101.88

TABLE 9

ANALYSIS OF SAMPLES (IN PERCENT) AFTER 17-1/2 HOURS  
AT 350° F AND 1,800 PARTS PER MILLION SO<sub>2</sub>

Component	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	A <sub>5</sub>
Fe <sub>2</sub> O <sub>3</sub>	11.2	12.8	6.9	6.4	7.8
TiO <sub>2</sub>	0.9	0.7	1.0	0.7	0.9
CaO	25.9	29.8	16.9	35.7	27.6
K <sub>2</sub> O	0.4	0.3	0.6	0.4	0.9
SO <sub>3</sub>	6.5	4.8	1.5	4.8	1.8
P <sub>2</sub> O <sub>5</sub>	0.8	1.0	0.5	1.2	0.9
SiO <sub>2</sub>	26.3	26.9	46.0	25.0	35.2
Al <sub>2</sub> O <sub>3</sub>	13.8	14.1	16.5	11.2	16.2
MgO	7.9	7.8	4.8	7.5	10.2
Na <sub>2</sub> O	5.1	2.0	5.3	4.3	0.2
Total	98.8	100.2	100.0	97.2	101.7



TABLE 12

ANALYSIS OF SAMPLES (IN PERCENT) AFTER 17-1/2 HOURS  
AT 800° F AND 1,800 PARTS PER MILLION SO<sub>2</sub>

Component	A1	A2	A3	A4	A5
Fe <sub>2</sub> O <sub>3</sub>	10.7	12.2	6.9	6.3	7.8
TiO <sub>2</sub>	0.8	0.7	0.9	0.7	0.8
CaO	25.6	29.5	17.1	34.7	27.3
K <sub>2</sub> O	0.4	0.4	0.7	0.4	0.8
SO <sub>3</sub>	9.2	6.0	2.7	7.5	2.0
P <sub>2</sub> O <sub>5</sub>	0.8	1.0	0.5	1.2	0.9
SiO <sub>2</sub>	25.2	26.6	45.2	23.9	34.7
Al <sub>2</sub> O <sub>3</sub>	13.9	14.4	16.7	11.0	16.4
MgO	7.8	7.9	4.8	7.2	9.8
Na <sub>2</sub> O	5.3	2.1	5.2	4.7	0.2
Total	99.7	100.8	100.7	97.6	100.7

TABLE 13

ANALYSIS OF SAMPLES (IN PERCENT) AFTER 17-1/2 HOURS  
AT 1,100° F AND 600 PARTS PER MILLION SO<sub>2</sub>

Component	A1	A2	A3	A4	A5
Fe <sub>2</sub> O <sub>3</sub>	10.6	12.0	6.9	6.2	7.7
TiO <sub>2</sub>	0.7	0.7	0.8	0.6	0.7
CaO	24.5	28.0	16.7	34.0	26.7
K <sub>2</sub> O	0.4	0.4	0.6	0.4	0.8
SO <sub>3</sub>	12.4	8.6	4.5	10.5	4.9
P <sub>2</sub> O <sub>5</sub>	0.8	0.9	0.2	0.7	1.0
SiO <sub>2</sub>	25.3	26.5	44.2	23.6	33.0
Al <sub>2</sub> O <sub>3</sub>	13.5	13.9	16.2	10.6	16.4
MgO	7.4	7.6	4.7	7.0	9.6
Na <sub>2</sub> O	6.0	2.7	6.2	5.0	0.4
Total	101.6	101.3	101.0	98.6	101.2

TABLE 16

ANALYSIS OF SAMPLES (IN PERCENT) AFTER 17-1/2 HOURS  
AT 1,600° F AND 600 PARTS PER MILLION SO<sub>2</sub>

Component	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	A <sub>5</sub>
Fe <sub>2</sub> O <sub>3</sub>	10.4	12.8	6.9	6.6	7.4
TiO <sub>2</sub>	0.4	0.4	0.4	0.3	0.5
CaO	24.8	29.2	17.5	36.0	26.2
K <sub>2</sub> O	0.3	0.4	0.8	0.4	0.9
SO <sub>3</sub>	12.7	6.6	3.4	8.1	6.8
P <sub>2</sub> O <sub>5</sub>	0.7	0.9	0.3	0.8	1.0
SiO <sub>2</sub>	23.2	25.6	44.1	22.2	33.3
Al <sub>2</sub> O <sub>3</sub>	13.4	14.3	16.3	11.2	15.6
MgO	7.9	8.6	5.1	9.0	9.7
Na <sub>2</sub> O	4.3	1.8	5.2	4.0	0.2
Total	98.1	100.6	100.0	98.6	101.6

TABLE 17

ANALYSIS OF SAMPLES (IN PERCENT) AFTER 17-1/2 HOURS  
AT 1,600° F AND 1,200 PARTS PER MILLION SO<sub>2</sub>

Component	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	A <sub>5</sub>
Fe <sub>2</sub> O <sub>3</sub>	9.0	11.1	6.6	6.3	7.1
TiO <sub>2</sub>	0.4	0.4	0.4	0.3	0.4
CaO	21.3	27.4	16.3	33.1	24.8
K <sub>2</sub> O	0.3	0.4	0.6	0.3	0.7
SO <sub>3</sub>	23.2	12.7	8.1	13.1	12.6
P <sub>2</sub> O <sub>5</sub>	0.7	1.0	0.2	0.7	0.9
SiO <sub>2</sub>	20.2	24.9	4.4	21.2	31.0
Al <sub>2</sub> O <sub>3</sub>	12.3	14.0	15.5	10.5	14.7
MgO	7.0	7.9	4.6	7.7	9.2
Na <sub>2</sub> O	4.1	1.7	4.9	3.9	0.2
Total	98.5	101.5	99.6	97.1	101.6

TABLE 20

COMPARISON OF SO<sub>2</sub> ABSORPTION OF FLY ASH SAMPLE NO. 4  
WITH ALKALIZED ALUMINA (PELLET AND CRUSHED)

Time, (hour)	SO <sub>3</sub> increase, percent		
	A4	CAA	PAA
0	0	0	0
1/2	5.8	0.5	0.1
1 1/2	6.4	1.1	0.9
3 1/2	7.1	0.9	1.7
7 1/2	9.9	4.1	4.2
17 1/2	11.9	13.4	12.0

Test conditions: 1,200 parts per million SO<sub>2</sub>  
1,100° F

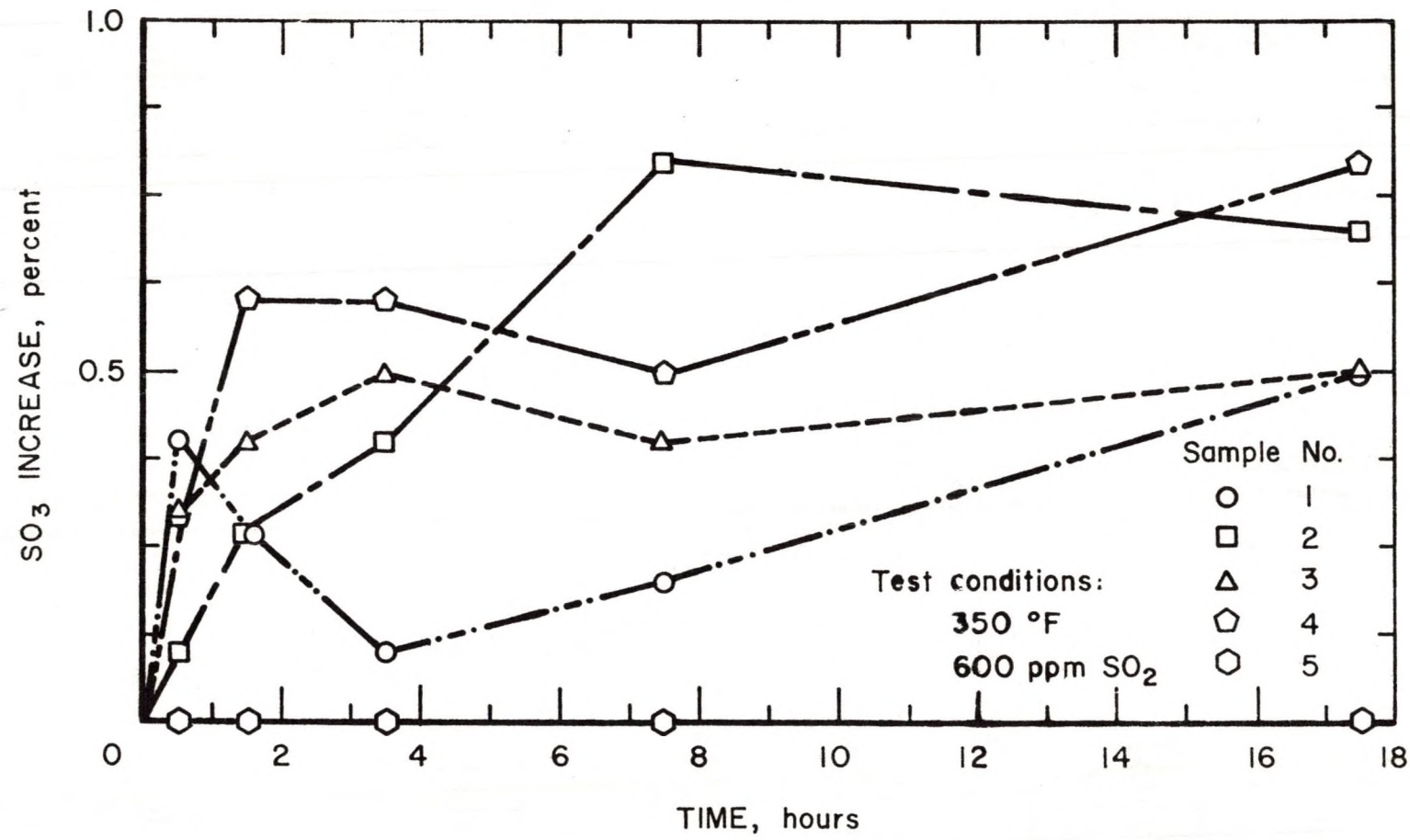


Fig 4 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (350 °F and 600 ppm SO<sub>2</sub>).

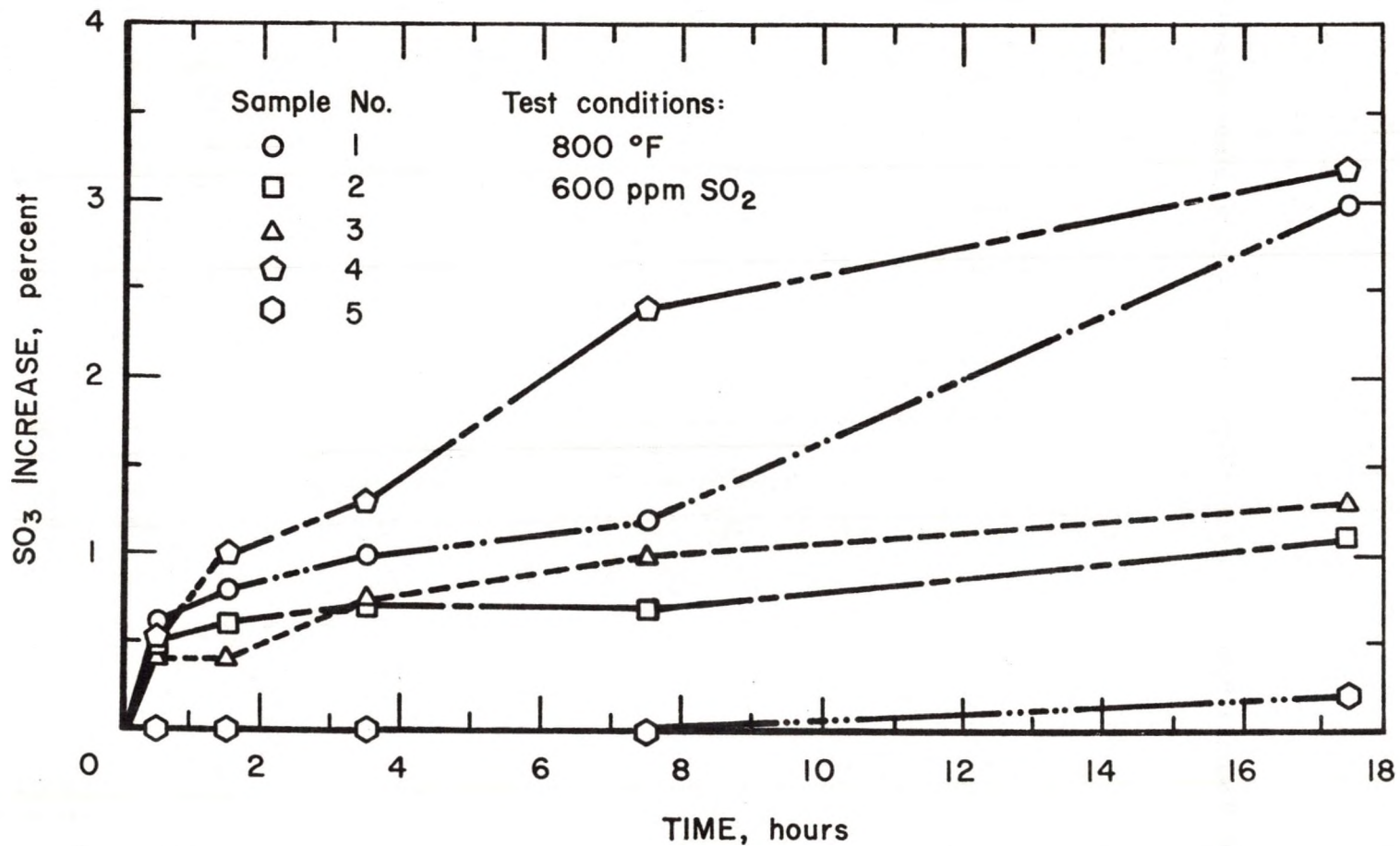


Fig 5 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (800 °F and 600 ppm SO<sub>2</sub>).

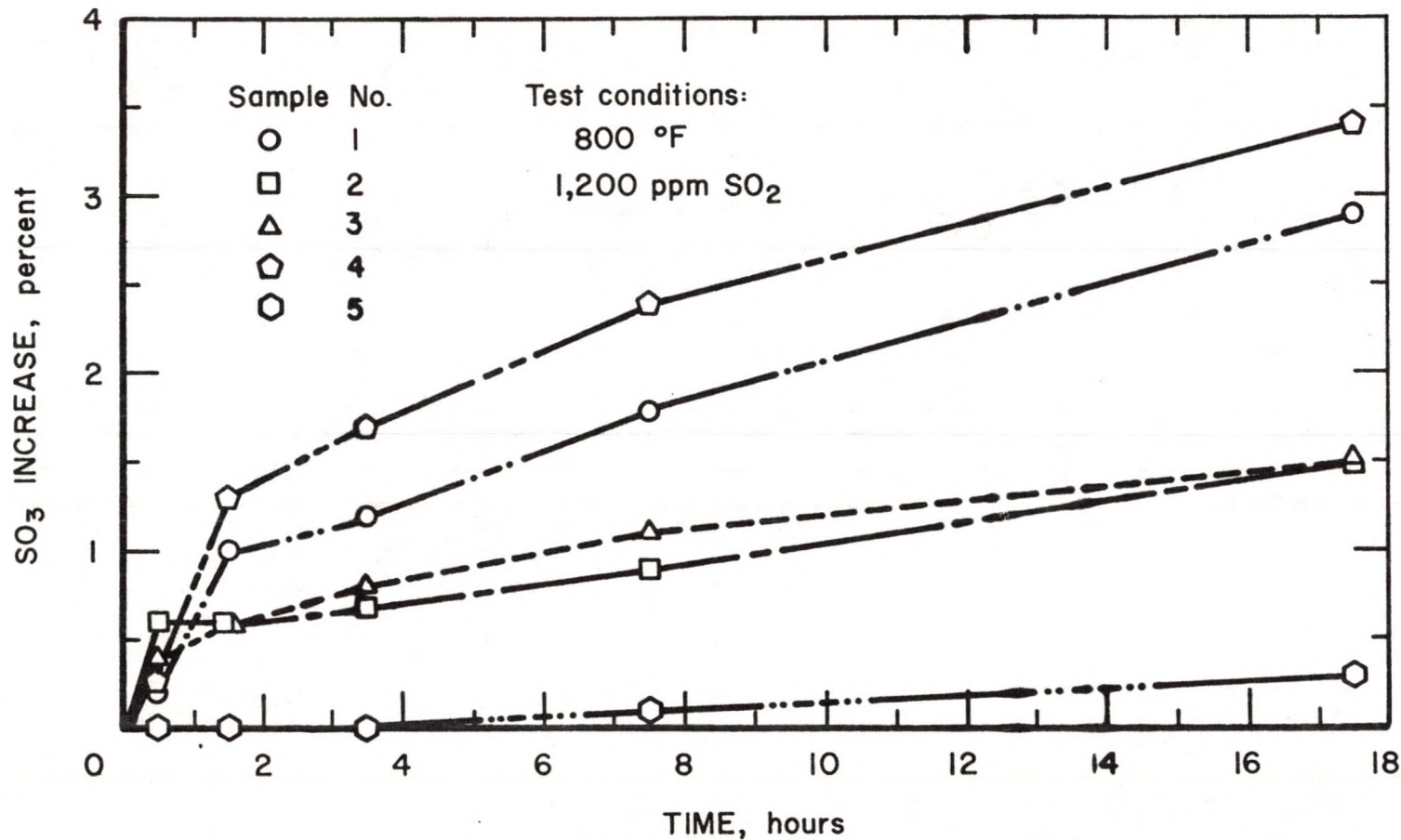


Fig 6 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (800 °F and 1,200 ppm SO<sub>2</sub>).

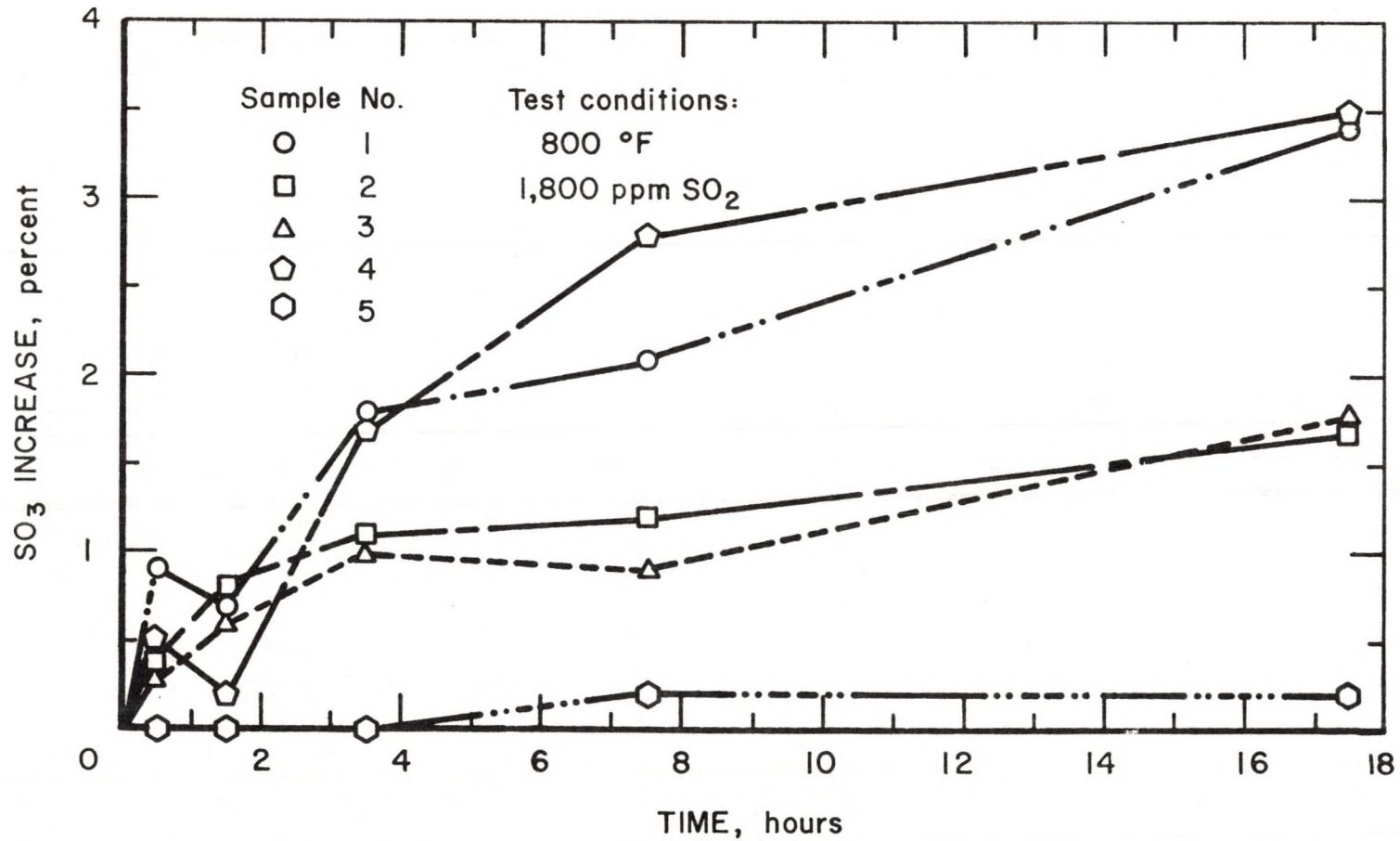


Fig 7 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (800 °F and 1,800 ppm SO<sub>2</sub>).

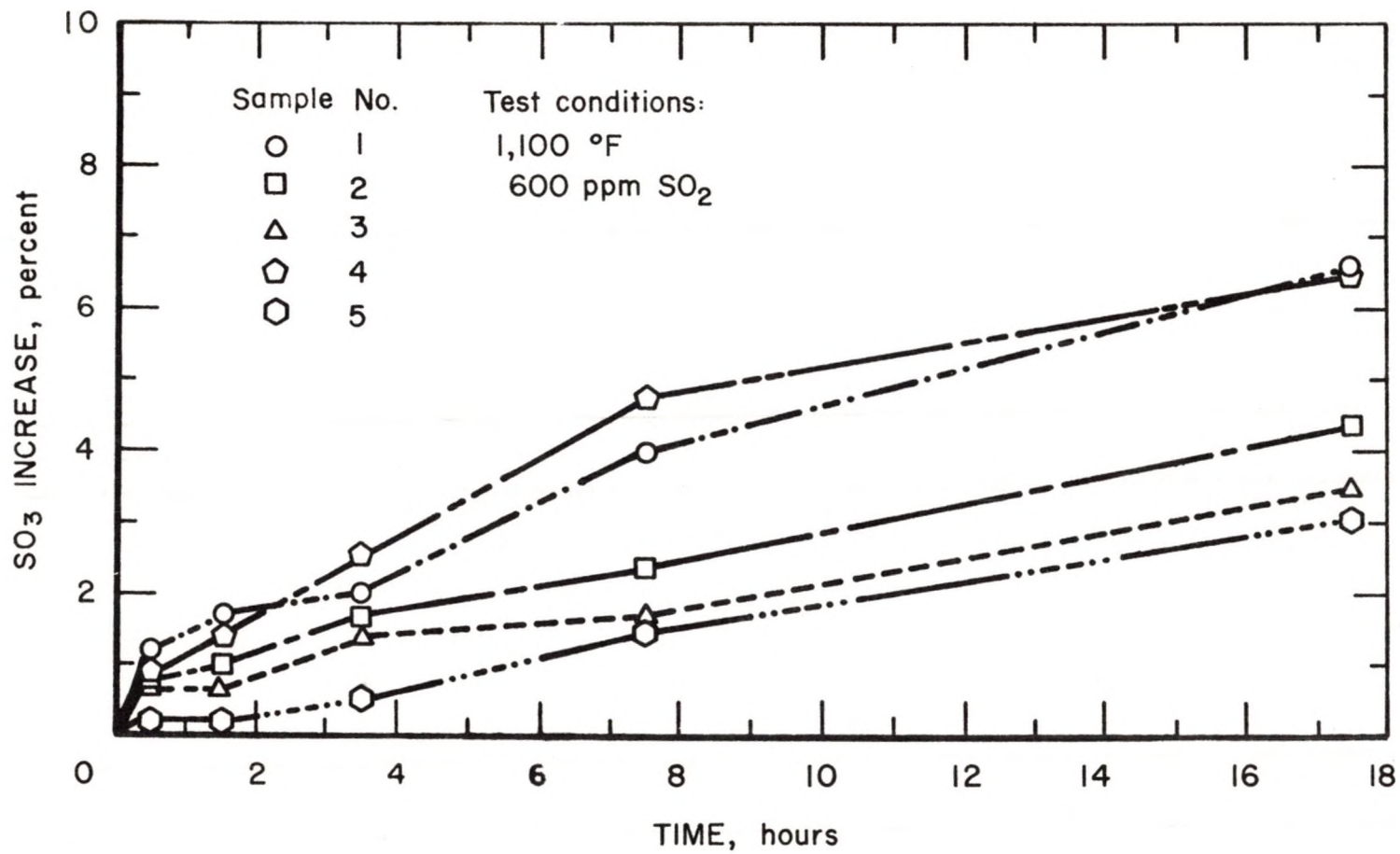


Fig 8 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (1,100 °F and 600 ppm SO<sub>2</sub>).



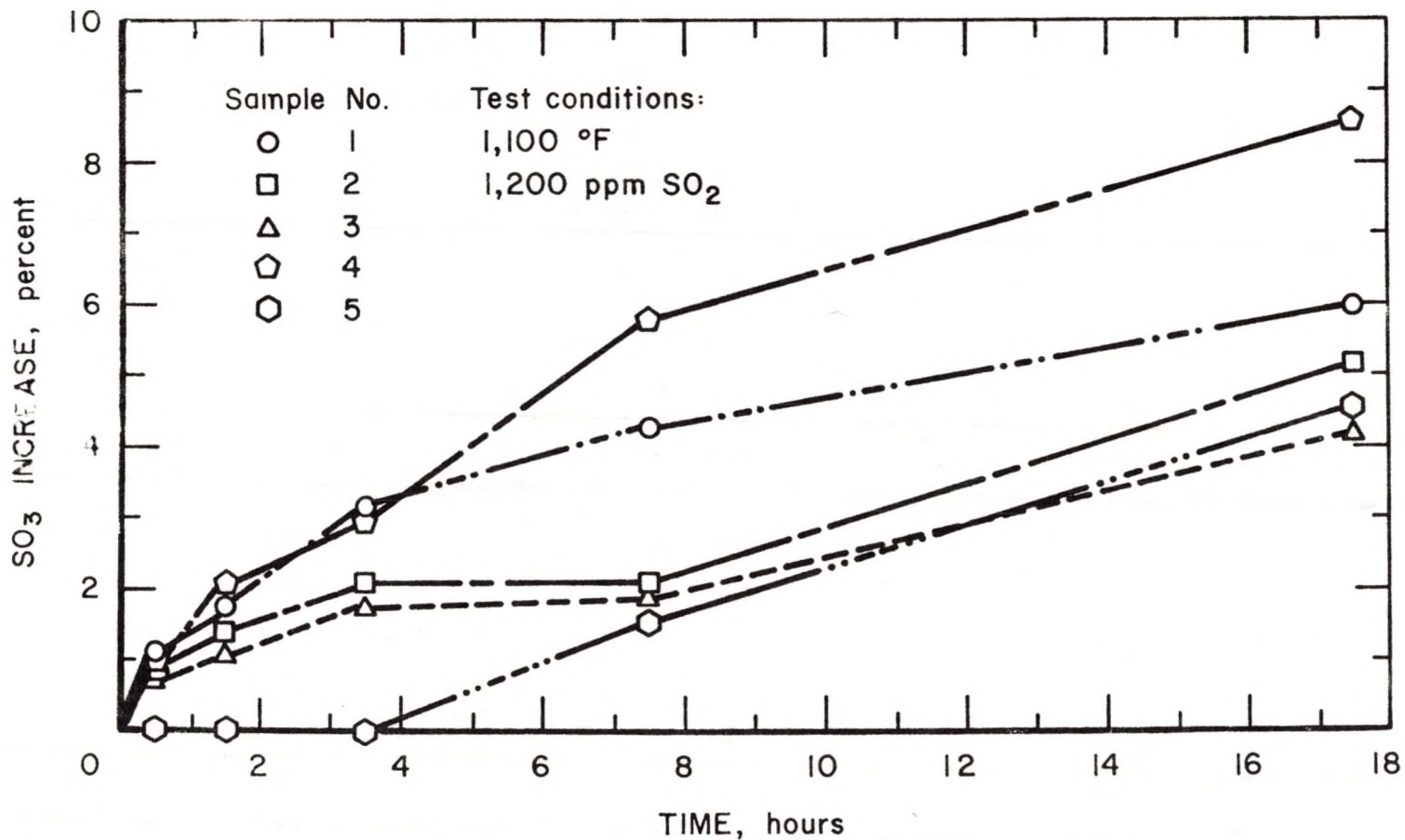


Fig 9 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (1,100 °F and 1,200 ppm SO<sub>2</sub>).

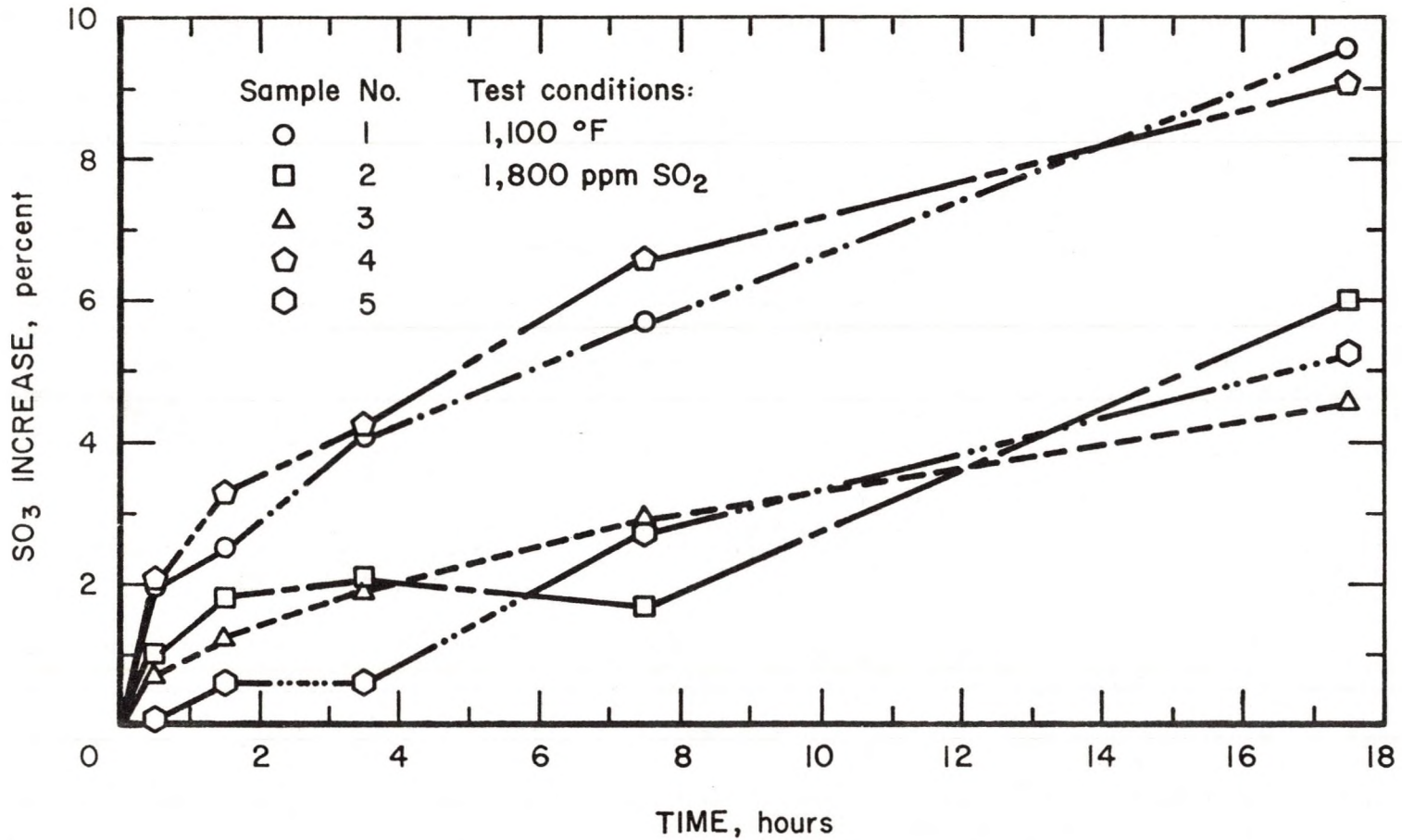


Fig 10 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (1,100 °F and 1,800 ppm SO<sub>2</sub>).

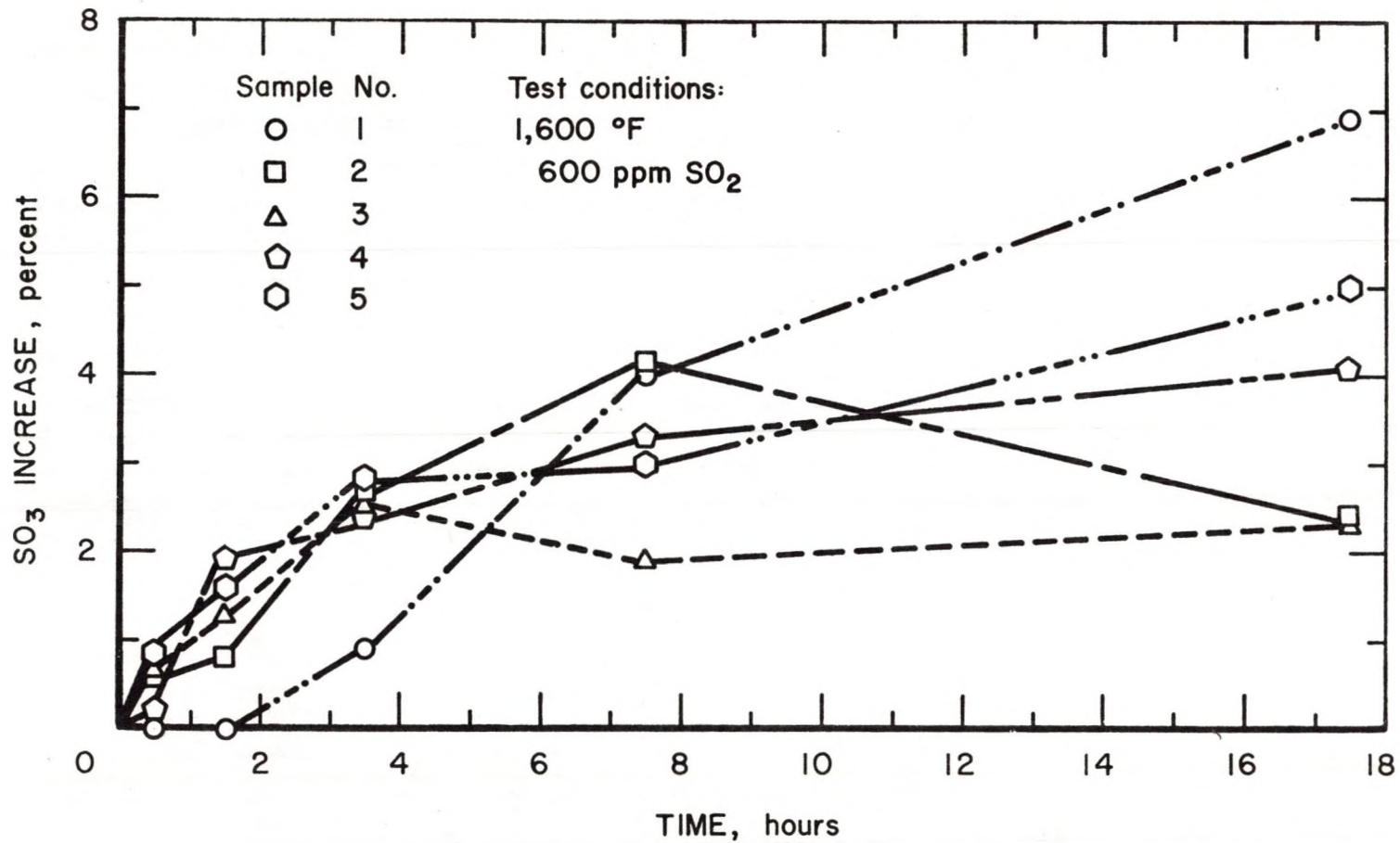


Fig II Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (1,600 °F and 600 ppm SO<sub>2</sub>).

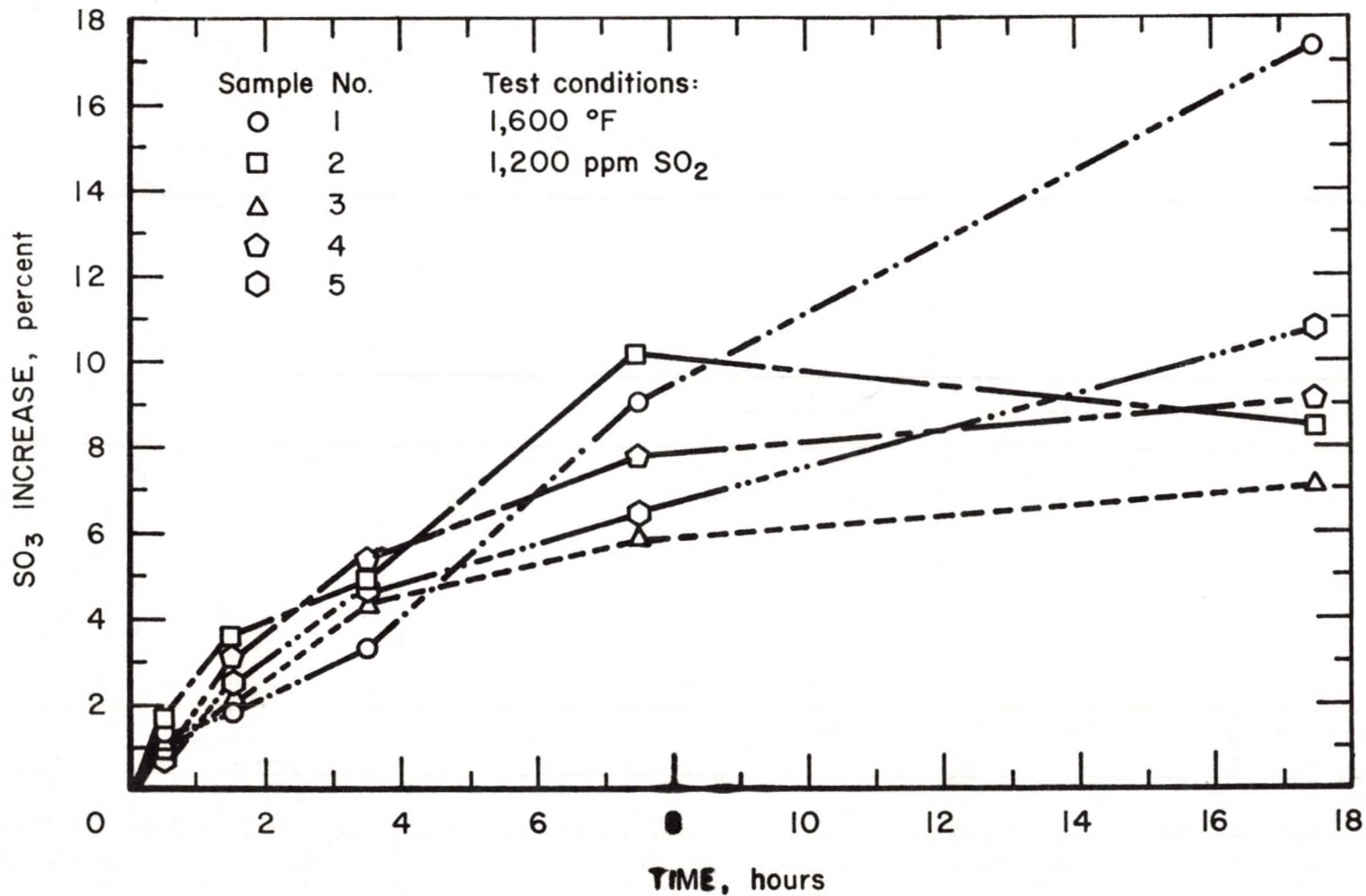


Fig 12 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (1,600 °F and 1,200 ppm SO<sub>2</sub>).

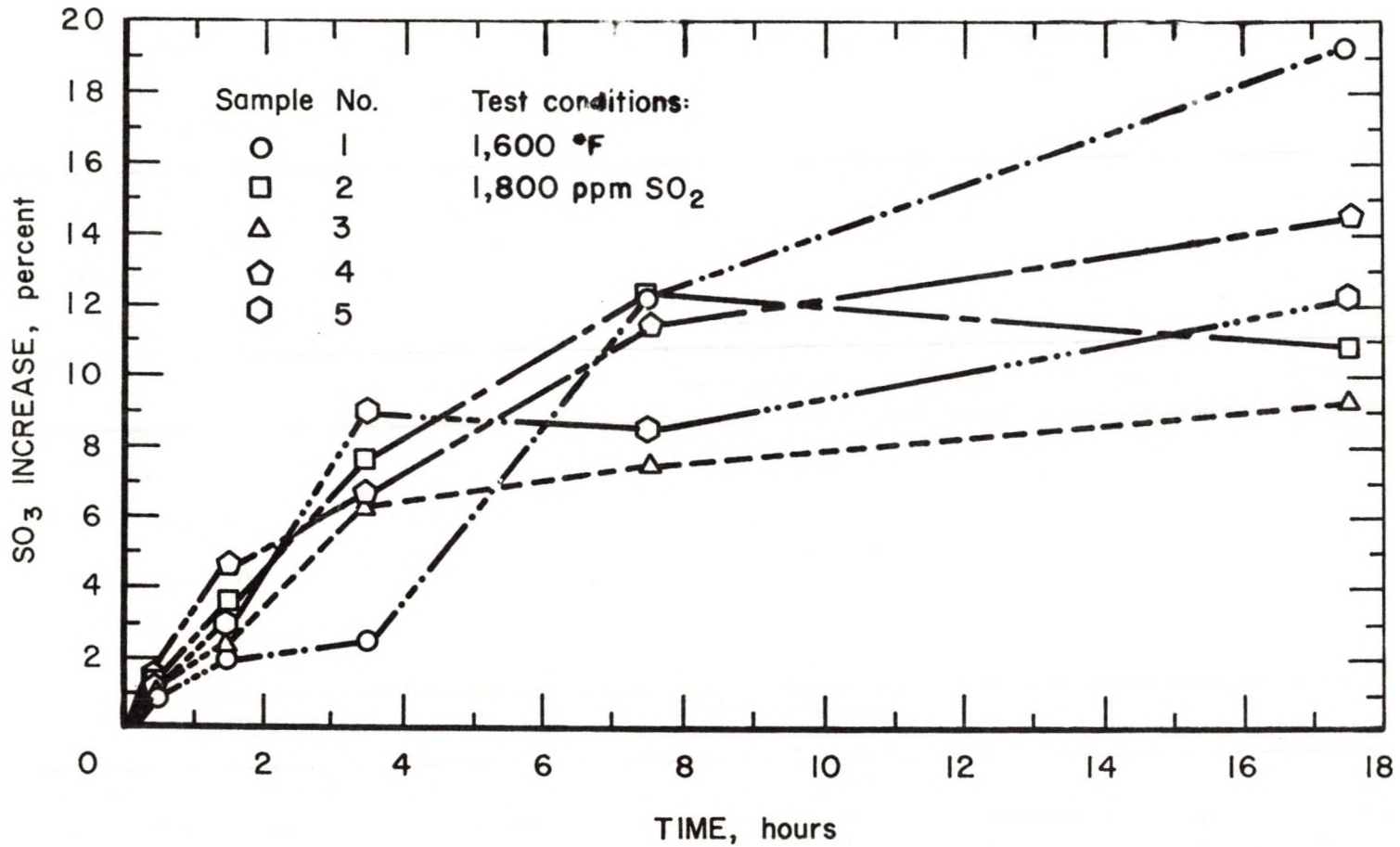


Fig 13 Percent increase in SO<sub>3</sub> versus time using five different samples of flyash (1,600 °F and 1,800 ppm SO<sub>2</sub>).

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APPENDIX C

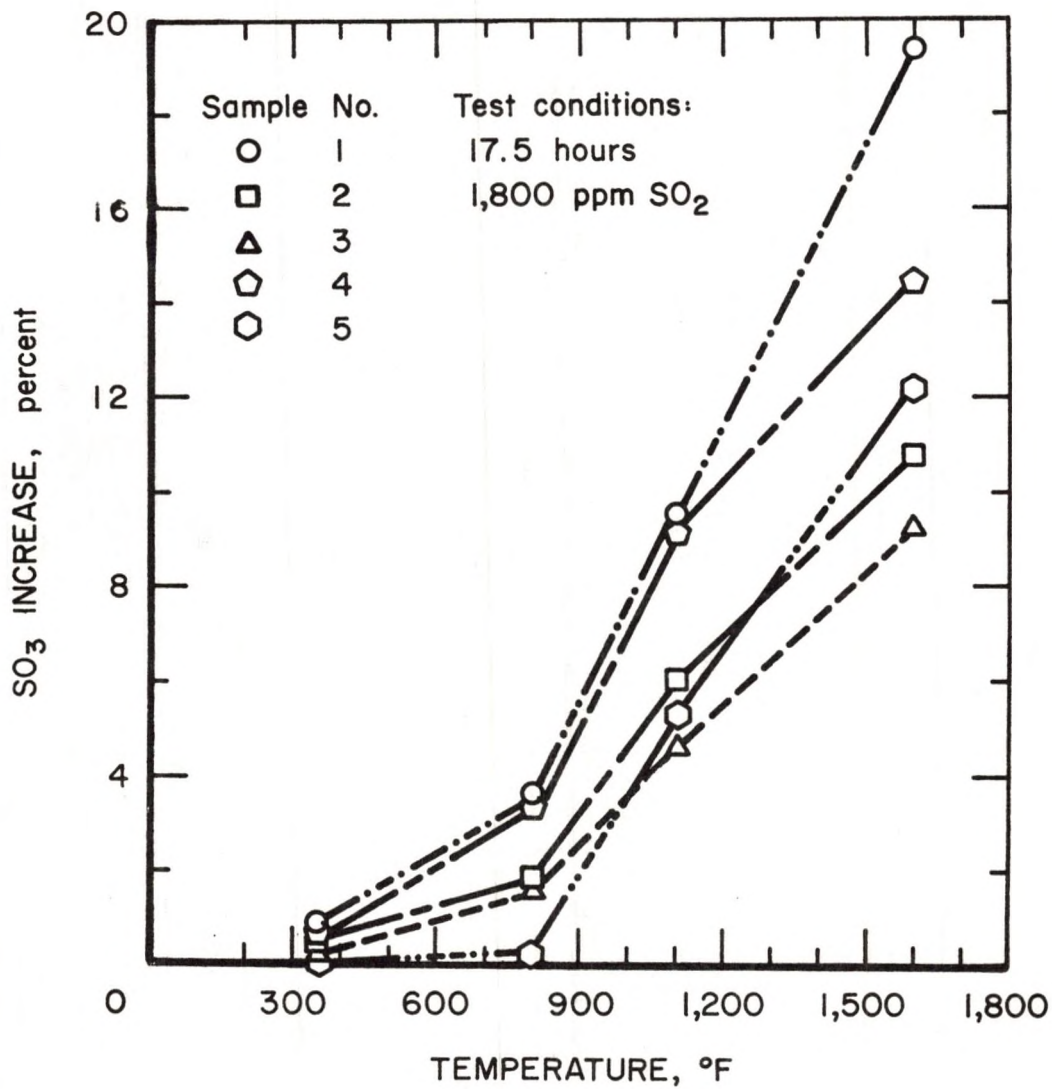


Fig 14 SO<sub>3</sub> in flyash after exposure to synthetic flue gas at various temperatures.

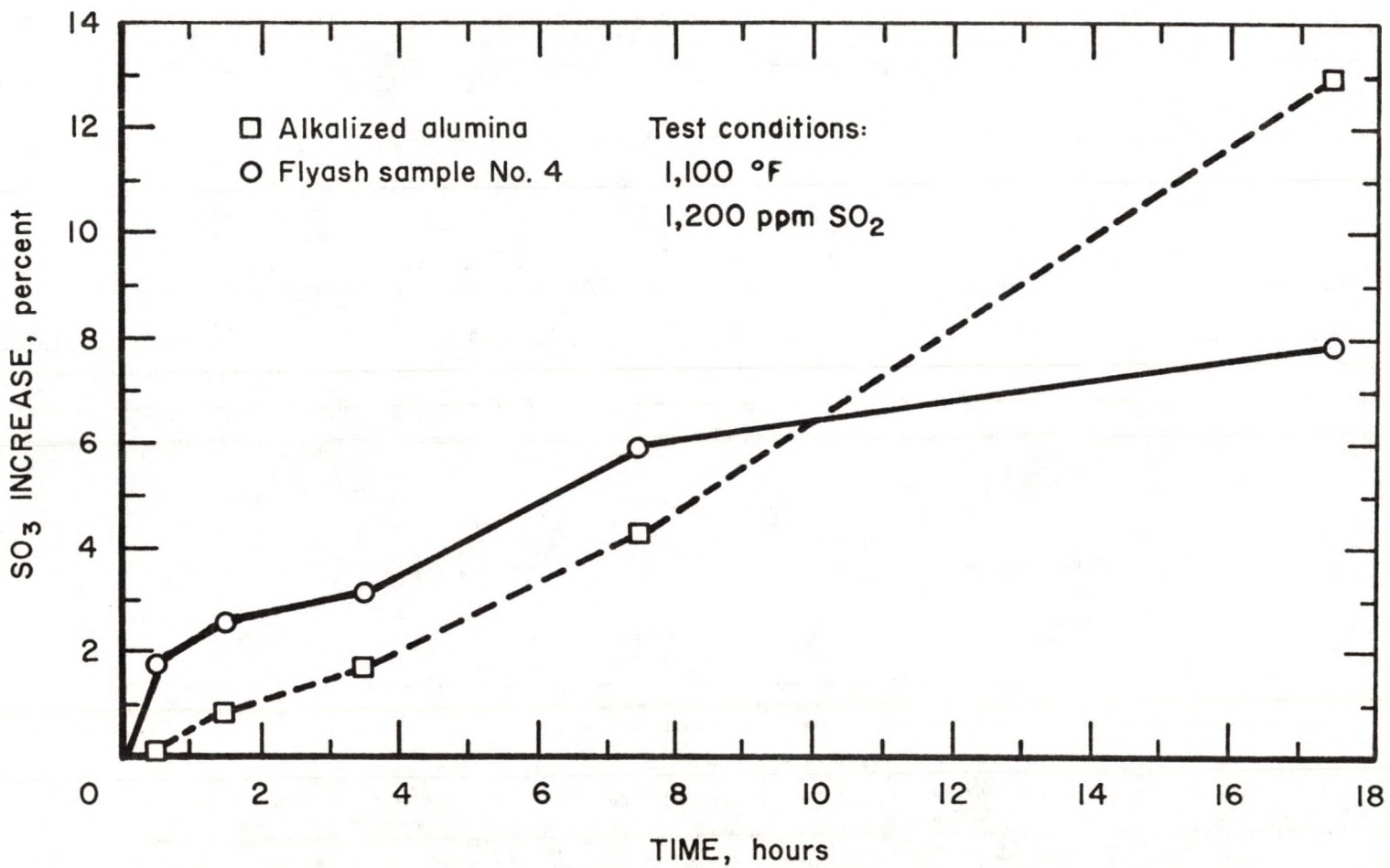


Fig 15 Comparison of flyash sample number four to alkalized alumina.

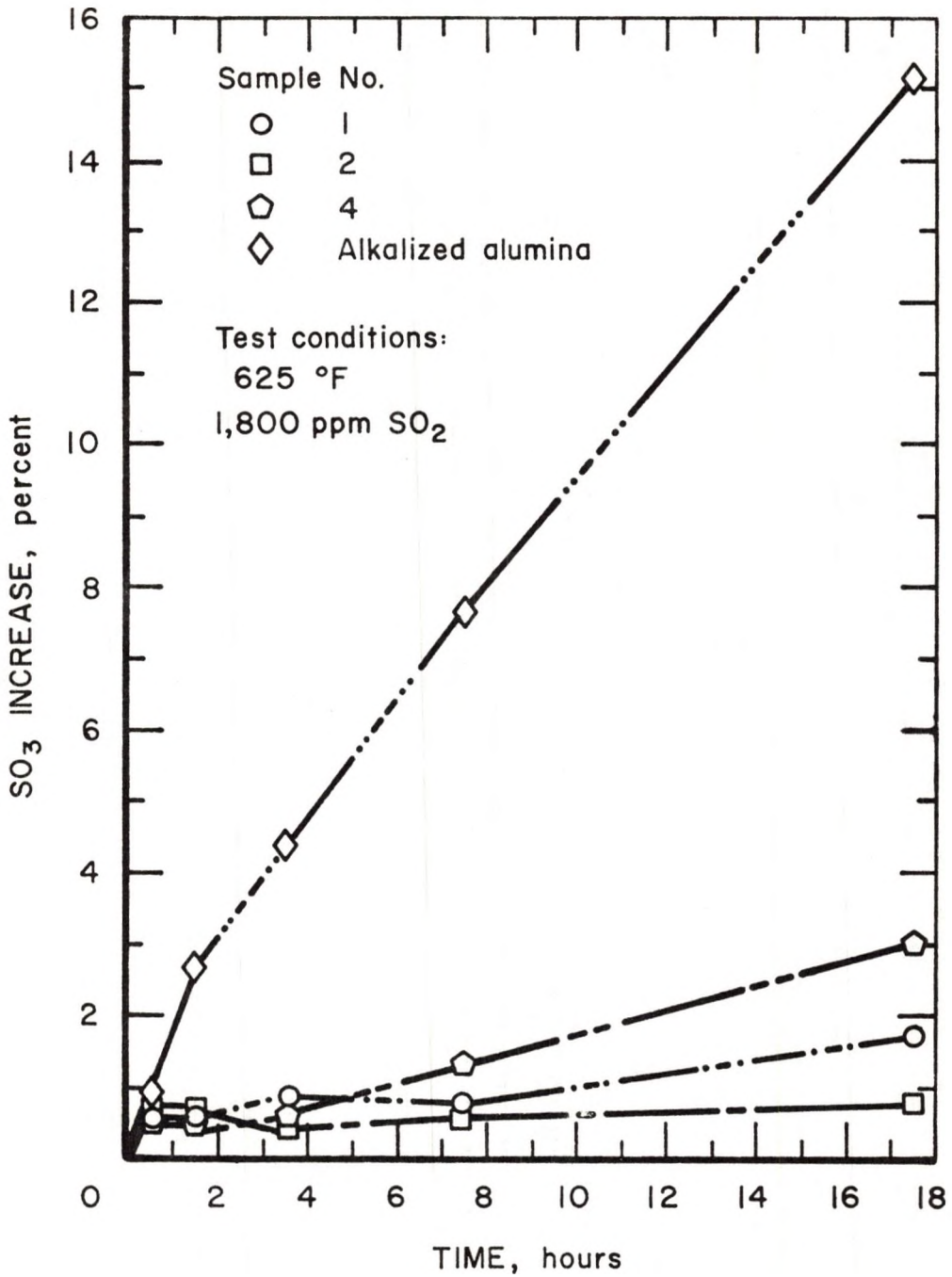


Fig 16 Comparison of three different samples of flyash to alkalized alumina.



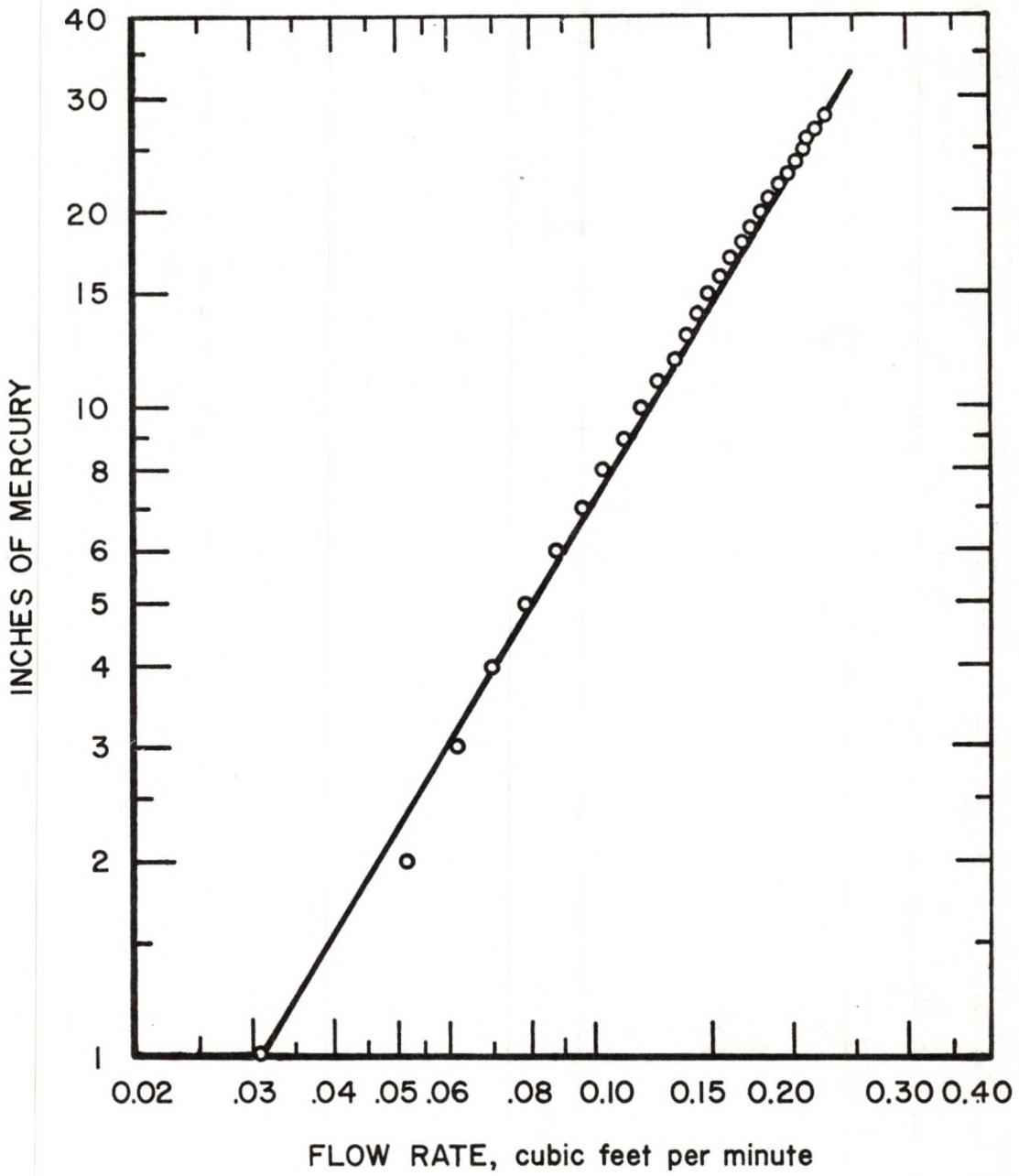


Fig 17 Calibration of air capillary.

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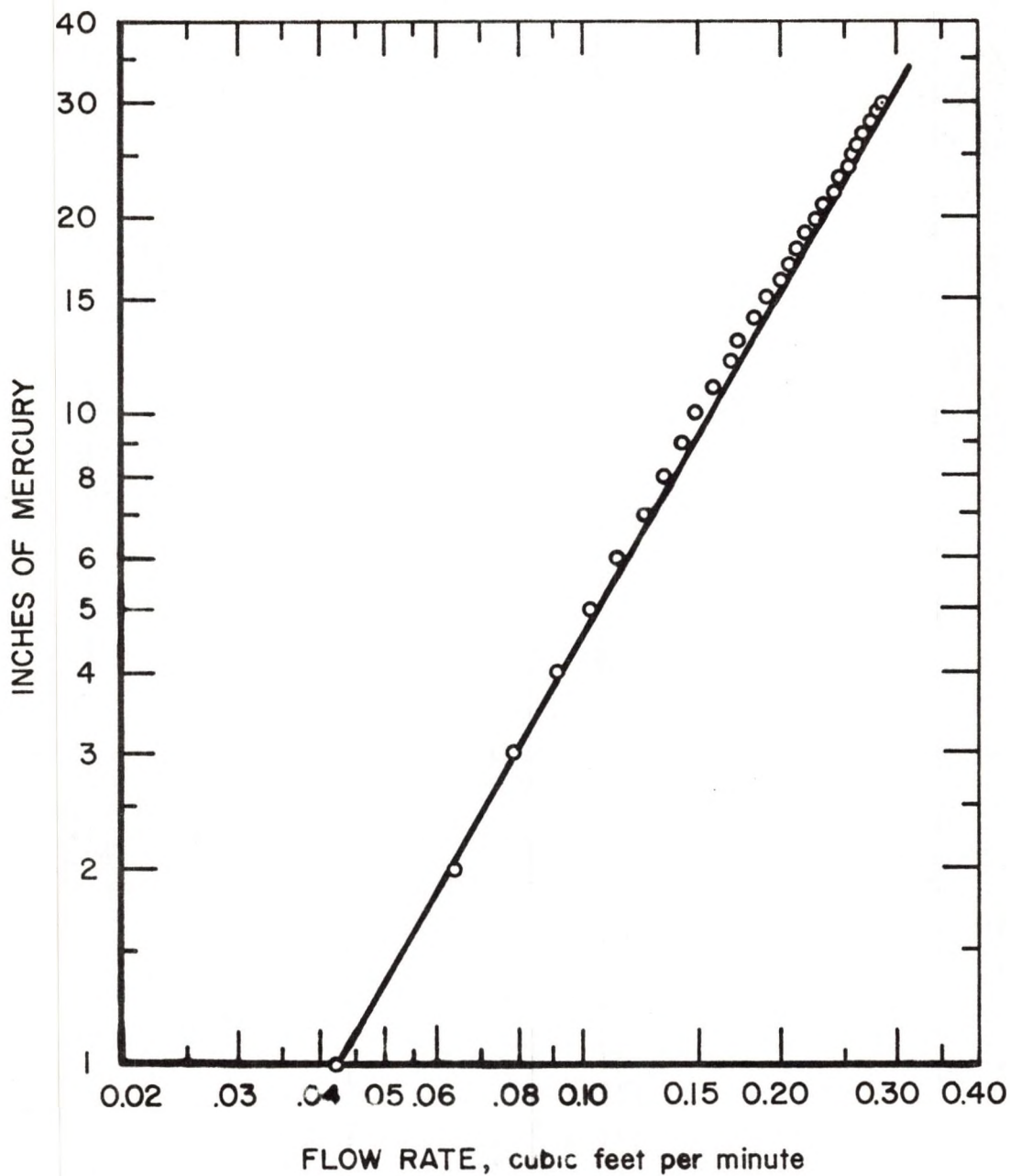


Fig 18 Calibration of nitrogen capillary.

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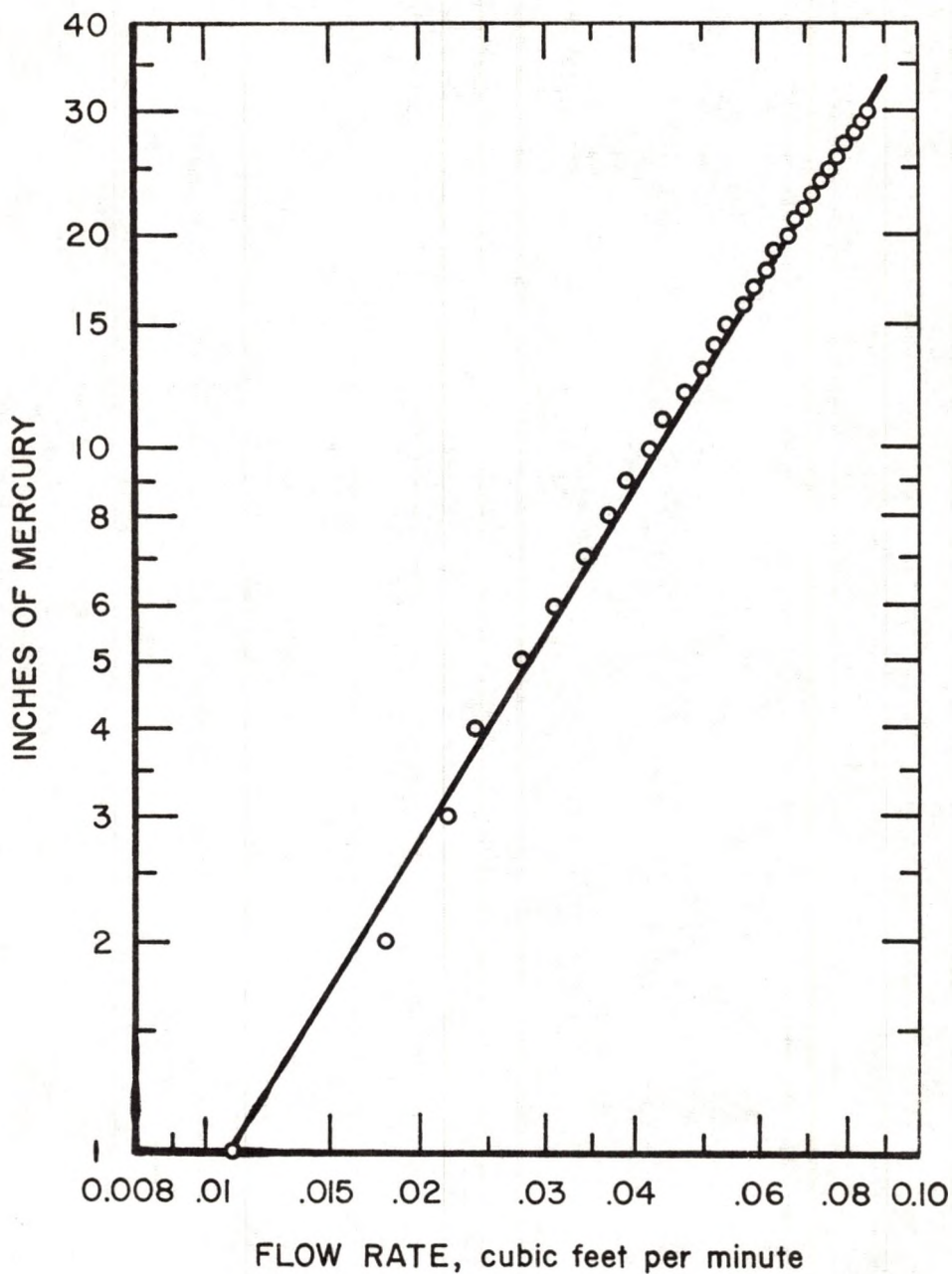


Fig 19 Calibration of carbon dioxide capillary.

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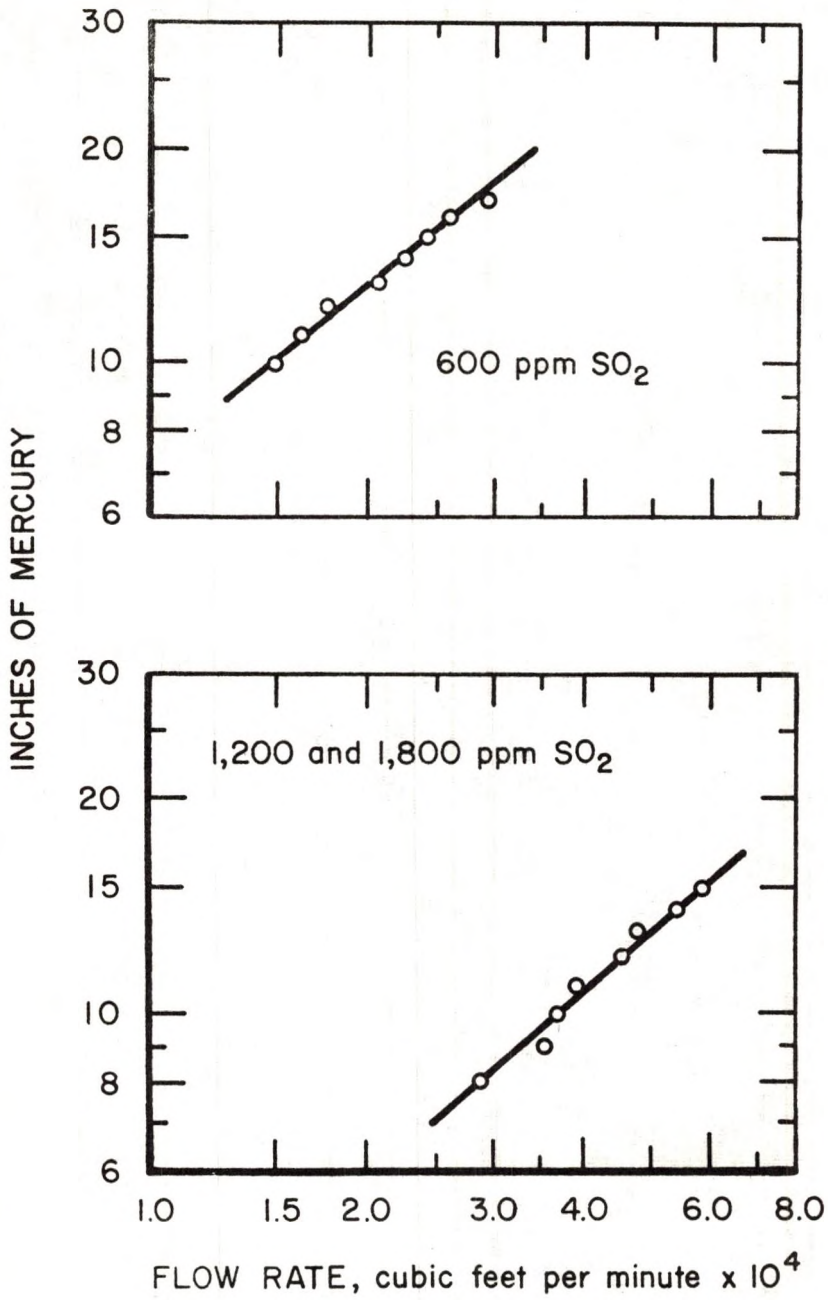


Fig 20 Calibration of SO<sub>2</sub> capillaries.

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