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SIMPLE ANALYTICAL MODELS FOR SULPHUR RETENTION IN FLUIDIZED BED COMBUSTORS

J.Q. Zhang, E.J. Anthony, V.V. Razbin and F.D. Friedrich

DIVISION REPORT ERL 89-28 (TR)

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SIMPLE ANALYTICAL MODELS FOR SULPHUR RETENTION IN FLUIDIZED BED COMBUSTORS

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ABSTRACT

Three one-phase analytical models have been developed to interpret sulphur retention data from three pilot-scale bubbling fluidized-bed combustors and Canada's first industrial fluidized bed heating plant. One is a well-mixed flow model. Two are plug flow models, one of which assumes that sulphur from coal is released at the base of the bed and the other assumes that the sulphur is released uniformly throughout the bed.

The SO_2/CaO reaction is assumed to be first order. Its overall rate is determined from data obtained in a bench-scale reactor. A modified pore plugging model is proposed to correlate the overall rate to the fraction of sorbent sulphated. This results in explicit solutions of the models.

The plug flow model which assumes that sulphur is released at the base of the bed agrees well with experiments when high volatile bituminous coal is fed underbed. Data were obtained for the in situ SO_2 concentration profiles measured in a 1 m^2 pilot plant and tests conducted at two other 0.155 m^2 pilot plants. The well-mixed flow model, however, underpredicts sulphur retention performance for the industrial heating plant when the coal is fed overbed. It is thought that this is due to the high fluidizing velocity employed, above 3 m/s, for the most of the tests.

The effects of recycle, bed overflow system and gas velocity on the effective size for the in-bed sorbent are discussed. The models indicate that no role is played by the volume occupied by the in-bed heat exchange tubes on the sulphur capture. The effect of the splash zone is also included in the models. The sulphur retention increases primarily with the Ca/S molar ratio and also with gas residence time, solids residence time and the feeding rate of the sulphur per unit volume of the bed.

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NOTATION

A	nominal cross-sectional area of the bed, m^2
a	empirical parameter for overall rate correlation, S^{-1}
b	reaction rate parameter, 1/s
Ca/S	calcium to sulphur molar ratio
C	SO_2 concentration, kmole/m^3
C_0	SO_2 concentration at base of the bed, kmole/m^3
C_h	SO_2 concentration at bed surface, kmole/m^3
D	maximum fractional conversion of the sorbent
F_{Ca}	feeding rate of calcium, kmole/s
F_s	feeding rate of sulphur kmole/s
g_c/g'	ratio of conversion factor to accelerator due to gravity
g	generation rate density of SO_2, kmole/m^3s
H	height of expanded bed, m
k_1, k_2	parameter in empirical overall rate correlation
M	total amount of sorbent in the bed, kmole
M_0	amount of sorbent per kg bed material, kmole/kg
m	mass efflux of solids product of the combustor, kg/s
m_b	total mass of in-bed solids, kg
n'	power, equation 3.1
n	power, varying between 0.5-3, equation 3.4
P	total pressure drop of the bed, kg/m^2
R	fractional sulphur retention
r	consumption rate density of SO_2, kmole/m^3s
t	exposure time, s
t_1	gas residence time, s
t_2	average residence time of the solids in the bed, s
U	superficial velocity, m/s
V	nominal volume of the bed, m^3
V_t	volume occupied by the in-bed heat exchange tubes, m^3
x	vertical distance from distributor plate, m
W	parameter defined by equation (4.8)

Greek symbols

α	sulphation level, fraction of the sorbent sulphated
ρ	molar density of calcium in the sorbent, kmole/m3

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INTRODUCTION

It is well recognized that sulphur emission levels are a key environmental constraint on the performance of coal-fired systems. Fluidized bed combustion (FBC) has the ability to reduce sulphur emissions in situ by adding limestone to the bed along with the coal and is also associated with inherently low NO_x emissions. FBC may therefore be regarded as one of the most promising technologies for utilization of high-sulphur fuels.

The development and application of FBC technology has been supported by Energy, Mines and Resources Canada since the seventies. A wide variety of Canadian coals have been tested under the auspices of CANMET in pilot-scale bubbling fluidized bed combustors located at the Combusion and Carbonization Research Laboratory (CCRL), Queen's University and Point Tupper, Nova Scotia. Test work has also been carried out at Canada's first industrial bubbling fluidized bed heating plant located at Summerside, Prince Edward Island.

The primary aim of this work was to re-examine the existing techniques and models for analysis and predicting of sulphur retention performance, and thus to identify the models or the type of models which best serve the purpose of estimating sulphur capture. A secondary aim was to develop an analytical model describing the physical and chemical processes of sulphur retention in bubbling fluidized bed combustors. We also attempted to analyze the effects of operating conditions on the sulphur retention to provide a guide for planning experiments, selecting sorbent and improving performance.

The sulphur retention performance in most of these trials were interpreted by pure empirical models (1,2,3,4). These models do not take into account the physical or chemical mechanisms of the process, and therefore lack general applicability. A comprehensive model has been developed at Queen's University (5). Although this model provides a fairly good description of the sulphur retention process in fluidized beds, it is too complicated for easy practical application.

An analytical model with simpler solutions developed by Argonne National Laboratory (ANL) (6) was also used to interpret some of the data obtained from an exhaustive assessment of different Canadian limestones for suitability as sorbents in FBC systems (7). However, to date, no systematic investigation has been conducted to obtain a simple, practical, analytical model of sulphur capture.

As part of a corrosion test program, measurements of in situ SO_2, O_2, CO, CO_2 and NO_x concentrations in the bed and freeboard of the Point Tupper fluidized bed testing facility were carried out (8). Results show that gas concentrations are strongly influenced by spatial location, both laterally and longitudinally, within the combustor. It is clear from this work that the gas moves upwards through the bed, in essentially plug flow, with little lateral mixing. This raises questions about the ANL model, which assumes a well mixed gas flow and uniform sulphur release throughout the bed, and encouraged a re-examination of existing sulphur capture models and development of a model which is consistent with data.

REACTOR MODELS

CATEGORIES OF MODELLING AND EXAMPLES OF REACTOR MODELS

The published models for sulphur capture fall into three general categories: comprehensive, semi-theoretical, and empirical.

Empirical models do not simulate physical and chemical processes occurring in the fluidized bed combustors and are severely constrained to specific test conditions. Examples of typical empirical models include the works conducted in China (9) and at CANMET (1-4).

In the comprehensive models, both the behaviour of the fluidized bed and the sorbent particle are considered. It has been found that major factors determining sulphur retention include:

1. Hydrodynamics in the bed which determines the gas mixing.
2. The release pattern of the sulphur content, which may be coupled with the devolatilization and combustion processes.
3. The mixing of solid particles.
4. The particle size distribution of the sorbent. (Small-size particles are usually more reactive.)
5. The attrition and elutriation of the sorbent. (Fine sorbent particles may be elutriated leading to a loss in retention efficiency, while attrition and breakage of the sorbent particles open fresh, reactive surface for sulphation and thus enhance sulphur retention.)

6. The sulphation rate of sorbent particles. (The SO_2 reacts with CaO rather than $CaCO_3$ because the calcination of limestone is much faster than sulphation. However, the reaction slows down progressively because a dense layer of $CaSO_4$ builds up on the walls of the pores inside the particles inhibiting the diffusion of SO_2.)

Because of the complex nature of the phenomena, a comprehensive model usually includes a set of equations containing a large number of parameters which have to be specified by empirical correlations or sometimes estimated theoretically. Although such models provide a good understanding of the sulphation process, they are too complex for practical application in design or data correlation. The so called plume model (10) for instance, belongs to this category.

Semi-theoretical models include the effects of physical and chemical mechanisms. However, to keep the models simple, the processes in the bed are not described in great detail. The fluidized bed combustor is usually treated as a reactor or a series of reactors with appropriate assumptions for solid mixing, flow pattern and the location of sulphur release. Conversely, the SO_2 sorbent reaction is determined by laboratory data from thermogravimetric analysis (TGA) or bench-scale fluidized bed reactors or empirical expressions based on these data.

These models therefore may be identified as reactor models. Analytical solutions for reactor models are relatively easy to obtain. The sulphur retention can be related to the calcium to sulphur molar ratio, Ca/S, and other operating parameters, by means of relatively simple expressions, which is to be preferred for practical applications.

Examples of reactor models are listed in Table 1. The ANL model (11) and the Westinghouse (Westinghouse Research Centre) models (12, 13) represent probably the most widely used and most satisfactory previous models in this category.

DESCRIPTION OF PHYSICAL PROCESSES IN REACTOR MODELS

A number of standard assumptions are made in modelling bubbling AFBC systems. The bed is generally assumed to be formed by one or two phases with well mixed flow or plug flow through these phases.

Two-phase models, which are often used to describe FBC systems, were developed specifically for beds of fine particles assuming fast rising bubbles. However, fluidized bed combustors normally employ particles of large sizes (0.5-1.5 mm) and in-bed heat exchange tubes tend to restrict the growth of gas bubbles. The bed is therefore more likely to be operated in a slow bubble regime (17,18).

In this regime, the interstitial gas velocity exceeds the bubble rising velocity and the fluidizing gas uses the bubble as a shortcut on its way through the bed. Consequently, there is little difference in gas concentration between the bubble and the emulsion phase. The bed can therefore be more closely approximated by a one-phase model. Significant variations in the in-bed gas concentrations in both vertical and lateral directions were found in the Point Tupper unit (8) strongly suggesting that a better representation of the real physical process in the bed would be the plume model (10). However, it is unlikely that such a complex description of the bed can be accommodated by a simple reactor model. The approximation of plug flow is thus preferred.

The patterns of solid mixing and SO_2 release are two other important aspects of the physical process that must be incorporated into a model. They both depend on how the characteristic reaction time compares with the time period required for a solid particle to travel a distance comparable to the dimensions of the bed. Experiments with solids tracer in a $1m^2$ bed, containing heat exchange tubes, show that it takes 10-30 s for the tracer particles to move throughout the bed (17). It takes much longer, usually several minutes, for limestone particles to be appreciably sulphated (7). Therefore, the generally accepted assumption of well mixed sorbent particles is reasonable.

The devolatilization time for coal particles is usually comparable to that for a particle to travel the characteristic length of the bed. For example, it takes only 20-30 s to complete the devolatilization for a 1/4" diameter particle of Eastern Canadian high-volatile bituminous coal such as Minto or Evans which contains 34-39% volatiles by weight (19). Since much of the sulphur content is contained in the volatiles, SO_2 release is a function of devolatilization time and the time for the subsequent combustion of the volatiles. The release of SO_2 is therefore not uniformly distributed throughout the bed. Two extreme cases have commonly been considered in the reactor models, i.e., uniform release throughout the bed or complete release at the base of the bed. There is evidence suggesting that the latter assumption leads to a better agreement between model prediction and experimental measurements (20).

INCORPORATION OF SO_2/CaO REACTION RATE DATA

It is generally accepted that the sulphur capture reaction is first order for SO_2 over the range of concentration of interest in FBC systems. The overall rate of sulphur capture changes appreciably with particle size of the sorbent. In order to simulate the SO_2/CaO reaction, both the size distribution of limestone particles and the rate data for the limestone over the size range distribution function are required. The situation may be further complicated by the processes of elutriation, attrition and breakage of the sorbent particles occurring in the bed, whose extent varies with operating conditions. Needless to say, a rigorous analysis of these processes is too complex for a semi-theoretical model. Simplification is usually achieved by defining an effective size for the in-bed sorbent particles, and using only the data for overall rate related to an effective sorbent size.

The overall rate of SO_2/CaO reaction incorporated in reactor models is normally derived from data obtained from TGA or bench-scale fluidized bed reactors. This probably is the only practical way to evaluate the reactivity of the sorbent. The measured sulphation rate constant determined for the same sulphation level, ℓ, which is defined as fraction of the sorbent sulphated, equivalent to that of the sorbent in the fluidized bed combustor, is then taken to predict the sulphur retention (12, 13). As will be discussed later, the sulphation level of the in-bed sorbent is itself related to the in-bed sulphur retention, due to the requirement of mass balance, and such a graphical methodology will fail to provide an explicit solution for the sulphation process and cannot be used to analyze the effects of operating conditions thoroughly.

A more appropriate way to deal with the overall sulphation rate data is to develop an empirical correlation based on these data and then incorporate it directly into the model. Almost all these correlations are expressed in terms of sulphation level of the sorbent varying as a function of the exposure time and the SO_2 concentration used. The sulphation rate d /dt or sulphation level for the in-bed limestone must then be obtained by integration of the expressions

$$\int_0^\alpha (d\alpha/dt)E(t)dt \qquad \text{or} \int_0^\alpha \alpha E(t)dt,$$

where E(t) is the residence time distribution function of solids in an ideal continuously stirred tank reactor (21) and

$$E(t)=(1/t_2)\exp(-t/t_2) \tag{2.1}$$

where t_2 represents the average residence time of the solids. Since sulphation rate also depends on SO_2 concentration, such a treatment is convenient only when SO_2 concentration is constant and thus need not be included in the integration. Also the time-dependent function must be simple enough to obtain an analytical solution for the integration. Therefore, application of these empirical expressions has been previously limited to well mixed flow models.

For plug flow models, a correlation for the overall rate of sulphation reaction, expressed as a function of sulphation level of the sorbent, is more desirable. Thus the above integration will be avoided entirely and the rate of the sulphation in the bed can be related to the sulphur capture in terms of the sulphation level of the in-bed sorbent as demonstrated later. This is also a more appropriate approach because the decrease in sulphation rate is basically caused by the accumulation of sulphated calcium on the walls of the pores inside the sorbent particles.

CONCLUSIONS ON SULPHATION MODELS

It appears that semi-theoretical models or reactor models are the most desirable for practical analysis of sulphur retention in fluidized bed combustors. Furthermore, plug flow for gas and well mixed for solids are likely to provide appropriate assumptions for the in-bed processes. The SO_2 reaction is first order for SO_2 and its overall rate can be determined from tests in TGA or bench-scale fluidized bed reactor. The rate constant correlations as functions of sulphation level of the sorbent are the most helpful in order to develop a plug flow model which will provide an explicit solution.

OVERALL RATE EXPRESSION FOR THE SO_2 SORBENT REACTION

The overall rate of SO_2 sorbent reaction was measured previously using a bench-scale fluidized bed reactor (7). For the present analysis, additional tests were carried out for Havelock limestone larger than 1 mm diameter. The reactor is made of two 53-mm I.D. stainless steel pipes, the lower and an upper sections being 350 mm and 1100 mm long, respectively, separated by a porous silica distributor. The lower pipe is filled with ceramic "saddles" and used as a preheater. The reactor is surrounded and uniformly heated by four pairs of 305-mm long half-shell resistance heaters rated at 1130 Watts each. Material is added to the reactor from a hopper by gravity and subsequently removed by a vacuum system. The elutriated dust is collected by an on line cyclone before the vacuum pump.

The bed material was "round-grained" pure silica sand with an average size of 0.4 mm and typically, 250 g was used in a test run. The minimum fluidizing velocity at 22°C was determined as 20 cm/s. At a superficial fluidizing velocity of 54 cm/s and temperature of 850°C the expanded bed height is about 27 cm during the tests. The composition of the synthetic flue gas employed was 5% O_2, 8% CO_2 and 0.28% SO_2, the balance being N_2. A paramagnetic analyzer for oxygen and IR analyzers for CO, CO_2 and SO_2 were used. (All of the analyzers could be connected to chart recorders as required.)

For each test, the system was fluidized with synthetic flue gas prior to adding any sorbent to the system. The SO_2 concentration in the synthetic gas through the reactor was then measured since it is typically 10% lower than that bypassing the reactor due to formation of SO_3. The equilibrium ratio of SO_3/SO_2 is 0.117 at 850°C and 5% O_2. In this work, the reaction rate of SO_3 with the sorbent is assumed the same as SO_2. The error due to the assumption is estimated at not more than a few per cent.

Limestone samples of 10 g were used for each run. A typical curve of the SO_2 concentration in the exhaust gas, for which the overall rate was calculated, is shown in Figure 1.

Most of empirical correlations for the overall rate of SO_2 sorbent reaction are expressed in terms of sulphation level of the sorbent as a function of exposure time. The most common form used is (6,7):

$$\alpha = k_1 \; 1-\exp(-k_2 t^{n'}) \qquad (3.1)$$

where k_1, k_2 and power n' are determined experimentally. However, the intrinsic kinetics of gas-solid reaction suggest that the decrease in the rate results from sulphated calcium accumulated on the walls of the pores inside the sorbent particles leading to an exponential decrease in the surface available for SO_2 capture (15,16). Upon mathematical manipulation, the overall rate can be shown to vary linearly with the sulphation level of the sorbent, i.e.

$$d\alpha/dt = C(b/\rho)\;(D-\alpha) \qquad (3.2)$$

where C is concentration of SO_2 in the gas and first order reaction for SO_2 concentration is assumed, D is the maximum fractional conversion that the sorbent can achieve at infinite time under specific testing conditions, b is a reaction rate parameter, ρ is molar density of calcium in the sorbent.

Our data indicate, however, that the overall rate decreases more rapidly for sorbent particles larger than 1.0 mm diameter. Most of the pores in the outer layer of these particles may be plugged before the sulphation of the entire particle ceases. Thus, while the reaction surface in the inner core still decreases following the pore plugging model, the SO_2 concentration available for the reaction, which may be characterized by the concentration at the boundary of the inner core, is reduced to a fraction of that in the ambient gas, i.e. Cf(t). Thus, the modified pore plugging model gives

$$d\alpha/dt = C(b/\rho)\;(D-\alpha)f(\alpha) \qquad (3.3)$$

where $f(\alpha)$ may be determined experimentally. Given the assumption of first order reaction, the empirical correlation (3.1) can be written as

$$d\alpha/dt = C(b/\rho)\;(D-\alpha)\;[-\ln(1-\alpha/D)]^n \qquad (3.4)$$

However, a more convenient form of this expression for particles smaller than 1.0 mm in diameter is

$$d\alpha/dt = C(b/\rho)(D-\alpha)^n \qquad (3.5)$$

When n' and n = 1, equations 3.1 and 3.5 reduce to an identical form, i.e. equation 3.2. When n = 2, equation 3.5 corresponds to another correlation previously reported (22), i.e.

$$\alpha/D = 1 - D/(D-at) \qquad (3.6)$$

where α is another coefficient determined by experiment.

Figure 1 shows typical results for the limestone for three different size fractions of Havelock limestone, from New Brunswick. The parameters in the power-law correlation for the 0.85-1.0 mm (18 x 20 mesh) sorbent shown in Figure 1 were averaged over three runs. Figures 1 and 2 indicate that both the reactivity and the ultimate conversion of the limestone, determined from the chemical analysis of the sorbent after tests, decrease with the particle size.

MODEL DEVELOPMENT

Our sulphur retention model is based on the following assumptions:

(1) The gas is formed by one isothermal phase, for which the SO_2 concentrations are the same in both the bubbles and the emulsion.

(2) The gas in the bed is in plug flow with no axial dispersion. The velocity of the gas does not vary with the vertical location in the bed despite the effects of the chemical processes such as devolatilization, combustion, calcination and sulphation on the total gas molar flowrate.

(3) The release of the sulphur from the coal is uniformly distributed at the base of the bed.

(4) The bed solids are well mixed.

(5) The sulphur capture performance of the in-bed sorbent with a wide particle size distribution is equivalent to the performance of the same amount of the sorbent with a specific size range which is defined as the effective sorbent particle size in the bed. The sulphur sorption rate is assumed to be the same as that determined in a thermogravimetric system or a bench-scale fluidized·bed reactor using the same sorbent at the same sulphation level as the average sulphation level in the fluidized bed combustor under the same conditions.

(6) The SO_2/CaO reaction is first order for SO_2 concentration. The overall rate of SO_2 sorbent reaction follows the modified pore plugging model and can be expressed as equation 3.3.

(7) Both the in-bed heat exchange tubes and the bubbles are assumed to be uniformly distributed in the bed.

For comparison, a plug flow model assuming a uniform release of sulphur throughout the bed and a well mixed flow model were developed. These two models are designated models 2 and 3 hereafter and the first model described above is designated model 1. For model 3, the gas is assumed to be well mixed rather than in plug flow, though it is required by assumption (2). For both models 2 and 3, assumption (3) is replaced by the assumption that the release of the sulphur from the fuel is uniformly distributed throughout the bed.

At steady state, given the use of the first two assumptions, mass balance for SO_2 requires that

$$UA(dC/dx)=(g-r)\ (A-V_t/H) \qquad (4.1)$$

where U is the superficial velocity, x is the vertical height above the base of the bed, A is the nominal cross-sectional area of the bed on which the superficial velocity is based, V_t is the volume occupied by the in-bed heat exchange tubes and H is the bed height. The SO_2 generation rate density g and its consumption rate density r due to sulphation reaction vary only with x following the assumptions (1), (2) and (3). Since the sorbent is well mixed and uniformly distributed, the sulphur capture rate varies in the bed only with the SO_2 concentration. Recalling assumptions (4) and (7), SO_2 consumption rate density can be expressed as

$$r = [M/(V-V_t)]C(b/\rho)\ (D-\alpha)f(\alpha) \tag{4.2}$$

where M is the total amount of the sorbent in the bed, t is its average sulphation level, V is the volume of the bed and

$$V = AH \tag{4.3}$$

In case SO_2 is released at the base of the bed, i.e., for model 1, g = 0. The concentration of SO_2 can be obtained by solving (4.1) and (4.2). The solution is

$$C = C_0\exp[-(M(b/\rho)\ (D-\alpha)f(\alpha)/UHA)x] \tag{4.4}$$

where C_0 is the concentration of SO_2 at the base of the bed, and

$$C_0 = F_s/UA \tag{4.5}$$

with F_s representing the molar feeding rate of sulphur. The sulphur retention R can be calculated by the expression

$$R = (C_0-C_h)/C_0 \tag{4.6}$$

where C_h is the concentration of SO_2 at the bed surface where x = H. Thus

$$R = 1-\exp(-w) \tag{4.7}$$

where

$$w = (MH/VU)\ (b/\rho)\ (D-\alpha)f(\alpha) \tag{4.8}$$

In the case that SO_2 is released uniformly throughout the bed, i.e. for model 2, g is a constant and

$$g = F_s/(V-V_t) \tag{4.9}$$

The general solution of (4.1) then has the form of

$$C = \exp[-wx/H]\ \int_0^x (F_s/UV)\exp[wx/H]dx \tag{4.10}$$

the value of w does not vary with the height within the combustor, integration gives

$$C = [F_s/M(b/\rho)\ (D-\alpha)f(\alpha))][1 - \exp[-wx/H]] \tag{4.11}$$

The sulphur retention is defined by

$$R = 1 - UC_h/\ \int_0^H gdx \tag{4.12}$$

which gives

$$R = 1 - [1-\exp(-w)]/w \tag{4.13}$$

In the case of well mixed flow, i.e. for model 3, the mass balance on sulphur gives

$$F_sR=MC(b/\rho)\ (D-\alpha)f(\alpha) \tag{4.14}$$

The SO_2 concentration C is a constant throughout the bed and equals to the concentration in the gas leaving the bed.
Therefore,

$$C = F_s (1-R)/(UA) \quad (4.15)$$

Equations (4.14) and (4.15) then lead to the following form for R

$$R = (1 + 1/w)^{-1} \quad (4.16)$$

It should be noticed that the overall rate of SO_2 sorbent reaction varies with the sulphur retention R, since the sulphation level of the in-bed sorbent is related to the sulphur capture by

$$\alpha = R/(Ca/S) \quad (4.17)$$

where (Ca/S) is calcium to sulphur molar ratio. Also, in a continuous operation, the total amount of sorbent in the bed can be related to the residence time of the sorbent and its feeding rate given the assumption of well mixed solids in the bed, i.e.

$$M=F_{Ca}t_2 \quad (4.18)$$

The feeding rate of the sorbent F_{Ca} can be expressed as

$$F_{Ca}=F_s(Ca/S) \quad (4.19)$$

By incorporating equations (4.17), (4.18) and (4.19) into equation (4.8), w may be expressed as

$$w = F_s t_1 t_2 (Ca/S) b(D-R/(Ca/S)) f(R/(Ca/S))/(V\rho) \quad (4.20)$$

where f (R/(Ca/S)) is a function of R/(Ca/S).

The basic forms of these three models, i.e. equations 4.7, 4.13 and 4.16 are similar to those of Westinghouse models and ANL model (11,12,13). Therefore, these models may be regarded as modified forms of the previous models. However, the present analysis shows no effect of the volume occupied by the in-bed heat exchange tubes. Also, in the present models, the sulphur retention can be related to operating conditions by a single equation because the overall rate of SO_2 sorbent reaction is expressed as a function of sulphation level.

These expressions reduce to much simpler forms if an ideal plug pore model applies, i.e., equation 3.2. Particularly, for models 1 and 3, equations 4.7 and 4.16 become

$$Ca/S = R/D - v\rho \ln(1-R)/(F_s t_1 t_2 bD) \quad (4.21)$$

$$Ca/S = (R/D) [1 + v\rho /[F_s t_1 t_2 b(1-R)]] \quad (4.22)$$

RESULTS AND DISCUSSION

VARIATION OF SO_2 CONCENTRATION WITH HEIGHT

The SO_2 concentration profile which results from in-bed physical processes such as the flow pattern and the location at which the SO_2 is generated strongly affects the sulphur retention. Therefore, comparison of model predictions with measured in-bed SO_2 concentration is essential to evaluate the model.

The three models predict quite different dependences for SO_2 concentration as a function of bed height for a given specific sulphur retention at specific operating conditions. For model 1, equations 4.4, 4.5 and 4.7 imply that

$$C = [F_s/(UA)](1-R)^{x/H} \qquad (5.1)$$

For model 2, equations 4.11 and 4.13 give that

$$C = [F_s/UAw][1 - \exp(-wx/H)] \qquad (5.2)$$

The SO_2 concentration is a constant in the case of well mixed flow, given by equation 4.15.

At the surface of the bed, the SO_2 concentration determined from these three models are the same. However, equation 5.1 predicts a SO_2 concentration profile which decreases with bed height, whereas equation 5.2 assumes an increasing SO_2 concentration with bed height.

Figure 3 compares these three model predictions for SO_2 concentration profiles as a function of bed height with the in situ SO_2 concentration profile measured in the 1 m^2 pilot-scale bubbling fluidized bed combustor. This unit is located at Point Tupper, Nova Scotia, and was used as part of an extended corrosion trial.

The measurements for gas concentrations were carried out by extractive gas sampling during the second last segment of a 1000 h duration of a 11,000 h corrosion test, sponsored by Energy, Mines and Resources Canada. The Point Tupper facility and the gas sampling program have been described previously in detail (8).

The location and labelling of the sampling ports are given in Figure 4. All the ports were located on the south side of the combustor except two, port W19 was located on the west side and port N21 was located on the north. The concentrations were determined at 250, 500 and 750 mm from the inside wall on which the sampling port was located. Two series of measurements were made at each port. The sampling gas was analyzed by means of a bank of Beckman analyzers for SO_2, CO_2, CO and NO_x and a Teledyne oxygen meter. The instantaneous concentration was then recorded every minute for a period of 10 minutes and the average of the 10 readings was taken as the determined concentration.

The results from ports A16, I16, A9 and I9 are not included in Figure 3 as these ports are very close to the walls and results may not be representative of the average values of SO_2 concentration. The results from port N21 were also excluded because the port was located too far from the bed.

Lingan coal was from the Sydney coal field, Nova Scotia. A chemical analysis of the fuel is given in Table 3. The limestone employed is quarried locally at Irish Cove and contains 93% $CaCO_3$.

The average operating conditions during the sampling program are given in Table 4. The SO_2 concentration at the surface of the bed was measured as 400 ppm. This gives an in-bed sulphur retention efficiency of approximately 84.2%.

The results illustrated in Figure 3 clearly show that the SO_2 concentration increases sharply at the location about 300 mm above the distributor plate and then falls gradually with bed height. Although the coal was fed only a few centimetres above the distributor plate, the SO_2 concentration profile is primarily a result of the devolatilization after entering the bed.

Although the measured SO_2 concentration profile does not agree completely with any one of these three models, there is no doubt that model 1 best represents the measured in situ SO_2 concentration. Model 2, the plug flow model assuming uniform release of SO_2 throughout the bed, is apparently the worst model in terms of describing the SO_2 concentration in the bed. Since it is essential for an analytical model to describe the in-bed SO_2 concentration profile correctly as mentioned previously, model 3 must be considered as a better model than model 2, even though plug flow provides a better representation for the hydrodynamics in the bed. The real values for sulphur retention are likely to be somewhere between the values predicted by models 1 and 3.

The foregoing discussion is based on the assumption that coal is fed at the bottom of the bed. The SO_2 concentration in the bed would be more uniform and model 1 would be expected to slightly overpredict the sulphur retention if coal was fed overbed. In order to simplify analysis, the models do not reflect the effect of the number and the location of coal feeders.

VARIATIONS OF SULPHUR RETENTION WITH OPERATING CONDITIONS

Equations 4.7, 4.13 and 4.16 show that sulphur retention time basically depends on three variables: the nominal gas residue time H/U which may also be interpreted as the contact time of the gas reactant with the sorbent; the amount of sorbent available for the reaction per unit volume of the bed M/V and the reactivity of the sorbent at the sulphation level α .

After relating w to the operating conditions, all these three models predict that sulphur retention depends on the reactivity of the limestone, calcium to sulphur molar ration, nominal gas residence time, solids residence time and sulphur feeding rate per unit volume of the bed, F_s/V. The product of the last three parameters, $q=F_s t_1 t_2/V$, has the dimension of kmole s/m^3 and does not vary significantly with the scale of the combustor, providing H does not significantly change with scale-up.

Thus, for example, in the trials conducted at the pilot plant installed at Queen's University, which will be described later, the parameter group q ranges from 0.50 to 1.84, whereas the values vary from 0.80 to 1.85 in the trials conducted at the industrial-scale fluidized bed heating plant located at Summerside, Prince Edward Island, which is also described later.

The calcium to sulphur molar ratio Ca/S plays the same role in affecting the variable MH/UV as the other operating parameters, i.e., F_s/V, t_1 and t_2. Moreover, higher Ca/S also reduces the sulphation level of the in-bed sorbent according to equation 4.17, which causes an increase in the overall SO_2 sorbent reaction rate. This makes calcium to sulphur molar ratio the most important operating parameter affecting the sulphur retention.

The volume occupied by the in-bed heat exchange tubes cancels out in the derivation. Thus the volume occupied by the in-bed heat exchange tubes plays no role in the calculation.

The sulphur retention in the freeboard usually counts about 10% of the total sulphur retention of the combustor and occurs mainly in the splash zone (11). The splash zone may be incorporated by an effective bed height to which all the same assumptions apply. As the bed height also cancels out in the solutions, and M includes the sorbent in the splash zone, the effect of the freeboard is automatically incorporated in the models.

Figure 5 shows the variation of sulphur retention with calcium to sulphur molar ratio predicted by these three models. In agreement with the previous discussion, the highest sulphur retention is predicted by model 1, whereas model 2 gives the lowest sulphur retention. The figure also shows that sulphur retention increases strongly as a function of the group q. Therefore, significant scatter is expected if the experimentally measured sulphur retention is only plotted against the calcium to sulphur molar ratio employed. The sorbent assumed to be used for the calculation in this figure is 0.85-1.0 mm (18 x 20 mesh) limestone from Havelock, New Brunswick, which contains 95% $CaCO_3$. Its reactivity is given by equation 3.5 with $b = 1240.2\ s^{-1}$, $D = 0.357$, $n = 1.83$ and $\rho = 24.47$ kmole/m^3.

COMPARISON WITH PILOT-SCALE TRIALS

Experiments were conducted at two pilot-scale bubbling fluidized bed combustors installed at the Combustion and Carbonization Research Laboratory (CCRL), CANMET and Queen's University. The apparatus at CCRL is shown schematically in Figure 6 and has been described in detail in an earlier report (23). The combustor is 4.8 m high and has the external dimensions of 0.94 x 0.97 m. The internal dimensions are 380 x 406 mm giving a bed area of 0.155 m^2. The distributor plate is fitted with 100 bubble caps to allow uniform dispersion of the fluidizing gas. Although only a few in-bed water cooling tubes were used for most of these tests the unit is designed to allow up to 48 horizontal in-bed tubes (21.3 mm O.D). The fuel and limestone were fed through a horizontal feed port located about 100 mm above the distributor plate. The flash in the flue gas was collected by a multicyclone containing three vaned collection tubes operated in parallel. The ash collected was reinjected into the bed as required. The bed is instrumented with thermocouples and pressure probes. The flue gas is analyzed continuously for concentrations of CO, CO_2, SO_2, NO_x and O_2. The SO_2 concentration was measured simultaneously by an IR and an UV analyzer for comparison. All measured data are recorded every 5 minutes by an automatic data acquisition/control system and stored on floppy discs by a computer. An electronic data logger also records all the data every 5 minutes as a back-up system.

The facility at Queen's University is essentially identical to that at CCRL, based on the same design. However, the rig at Queen's University is equipped with a bed overflow port located 1.19 m above the distributor plate, which determines the nominal bed height, whereas in the rig at CCRL, the bed material is drawn from a drain near the base of the bed when necessary to control the bed level.

Three Eastern Canadian bituminous coals, i.e., Minto, Evans and Brogan, were tested. The chemical analysis data for these coals and the operating conditions for these trials are given in Tables 3 and 5. More detailed information can be found in separate reports (3,24,25).

For the tests at Queen's University, the solids residence time was determined by

$$t_2 = m_b/\dot{m} \qquad (5.3)$$

where $\dot{m}$ is total mass efflux of solids product, or sum of the bed product flux, i.e. the cyclone product flux and the baghouse product flux, and is measured experimentally, m_b is the mass of the solids in the bed determined either from the total bed pressure drop ΔP assuming

$$m_b = A\Delta P\, g_c/g \qquad (5.4)$$

defined as the fraction of the cyclone ash that is returned to the combustor. The rest of the ash collected by the cylcone was drawn off as product through an adjustable splitter valve.

For the tests at CCRL, the bed material was weighed after each run. The amount of sorbent (in kmoles) per kg mass of the bed material, M_0 was determined from chemical analysis of the bed material samples. Hence

$$M = M_0 m_b \qquad (5.5)$$

The calculated sulphur retention is then compared with the experimental value averaged over the last two hours of operation.

For the tests conducted at Queen's University, the effective size of the in-bed sorbent was taken as the diameter at which the distribution function of the sorbent particle size, determined by sieve analysis before feeding to the combustor, equals 1/2, i.e., the diameter of the sieve opening that passes 50% by mass of the sorbent particle in a sample. The 0.6-0.85 mm (20 x 30 mesh) and 0.3-0.6 mm (30 x 50 mesh) Havelock limestone samples were taken to represent the 0.44-1.5 mm (11 x 34 mesh) and 0.15-0.99 mm (16 x 100 mesh) sorbent corresponding to the effective size of 0.78 mm and 0.49 mm, respectively. The overall sulphation rate of these two limestone samples was determined from experimental data at a bench-scale fluidized bed reactor expressed as power-law correlation (3.5) with $b = 686.1\ s^{-1}$, $D = 0.381$ and $n = 1.05$ for the 0.65-0.85 mm sample; $b = 434.3\ s^{-1}$, $D=0.418$ and $n = 0.912$ for the 0.3-0.6 mm sample, and $\rho=24.46$ kmole/m^3 for both. However, all three models failed to interpret the sulphur retention data from trials with Brogan coal and 0.85-2-36 mm (8 x 20 mesh) Havelock limestone at CCRL when the average size of the sorbent fed (1.55 mm) was assumed to be the effective size for the in-bed sorbent. The 0.85-1.0 mm (18 x 20 mesh) Havelock limestone sample was found to be more representative of the in-bed sorbent for the tests at CCRL.

In Figure 7, the measured sulphur retention data in the tests with 11 x 34 mesh Havelock limestone are compared with those predicted from the three models. A similar comparison is shown in Figure 8, but only model 1 was used. Figure 7 shows that models 1 and 3 tend to under-predict the sulphur capture, whereas model 1 gives the most satisfactory results. Both Figures 7 and 8 show that the measured data is usually less than 20%. The largest discrepancy occurs when a low Ca/S molar ratio was employed.

As mentioned previously, the actual sulphur retention is more likely to fall between the predictions of models 1 and 3. In order to further compare these two models, the Ca/S molar ratio required to achieve the measured sulphur retention was calculated assuming that the overall rate of sulphation varies linearly with the sulphation level of the sorbent and the overall rate data were refitted by equation 3.2. In Figure 9, the results are compared with the Ca/S molar ratio actually employed in the tests with both 11 x 34 mesh and 16 x 100 mesh Havelock limestone.

It appears that model 1 is a much better model than model 3 in predicting the Ca/S molar ratio. In particular, model 3 failed to give a reasonable Ca/S molar ratio when high sulphur retention was required. Typical examples of such results were shown when sulphur retentions of runs 830202, 830329 and 840124 were calcualted. Model 3 predicts that the Ca/S molar ratios of 8.0, 15.9 and 29.1 are needed to achieve sulphur retention of 97.5%, 989.4% and 99.2%, which is much higher than actually employed. Model 1 however, gives only slightly lower Ca/S ratios, on average 5.9% lower than actually seen for these three tests with a largest discrepancy being 16.3% lower than the experimental result. The difference between model 1 and model 3 or model 2 when high Ca/S molar ratios are required can also be seen from Figure 5.

Figure 10 shows that the sulphur retention in the trials with Brogan coal and 0.85-2.36 mm (8 x 20 mesh) Havelock limestone can be predicted by model 1 within an error range of 20% assuming an effective size of 0.85-1.0 mm (18 x 20 mesh) for the in-bed sorbent, although sieve analysis indicates only about 11% of the total population in the 8 x 20 mesh limestone passes the 18 mesh sieve. For sorbent particles with a large size, the sulphation reaction is virtually stopped by plugging of the pores in the outer layer of the particle preventing its interior from being sulphated. The sulphation capacity of these particles therefore is lower than small particles. However, physical processes such as breakage and attrition encountered by the large sorbent particles in the bed open fresh surface for sulphation and thus increase the sulphation capacity. It seems that the sorbent fines generated only play a secondary role in reducing the effective size of the sorbent. This is suggested by the fact that both the measured sulphur retention data, with and without recycle, can be fitted by the model over a reasonable error range using identical effective size for the sorbent.

The data plotted in Figure 10 also indicate that the sulphur retention tends to be under-predicted when a higher velocity is employed. High velocity (about 3 m/s) enhances the abrasion and breakage of the sorbent particles and therefore reduces the effective size for the in-bed sorbent. At very high velocity (> 3 m/s), both the size and the rising velocity of the bubbles increase and eventually a point is reached at which the combustor is no longer operated in the small bubble regime.

The SO_2 concentration in the bubbles will become considerably lower then that in the emulsion phase which determines the rate of sulphation reaction. The sulphur retention will then be under-predicted by these one-phase models which calculate the SO_2 concentration from the total gas flow without making any distinction between bubbles and the emulsion phase. However, this does not occur in most of these tests, since the model does not under-predict the sulphur retention in the trials with 11 x 34 mesh and 16 x 100 mesh Havelock limestone for which the same range of velocity was employed.

Figures 7 and 8 indicate no significiant effect of recycle on the sulphur retention, whereas Figure 10 shows the contrary. This probably results from the fact that the two pilot plant facilities employed different bed withdrawal systems. Since fine bed material tends to segregate and collect in the upper part of the bed, more limestone fines will be drawn out of the combustor by the bed overflow system used by the rig at Queen's University when the cyclone ash is reinjected back to the bed. The effect of recycle on sulphur retention is thus greatly reduced. In the trials with Brogan coal and 8 x 20 mesh limestone, the excess bed material was drawn from a drain near the base of the combustor. The recycle of the cyclone ash therefore helps to increase the utilization of the sorbent. Further examination of the data plotted in Figure 10 indicates that the effect of recycle is more significant, as might be expected, when higher fluidizing velocity is employed. For the three ranges of velocity employed, 1.3 m/s, 2.1 m/s and 2.9 m/s, the sulphur retention achieved with recycle increased by 6.1%, 11.3% and 15.4% in average compared with the trials without recycle.

The difficulty in determining the effective size for the in-bed sorbent when sorbent of large size is employed represents the most serious shortcoming of these reactor models. The problem is further complicated by the fact that the effective size may differ significantly from the average size of the sorbent and vary slightly, depending on the superficial velocity used and whether cyclone ash is recycled. This will be discussed later.

"OMPARISON WITH TRIALS AT SUMMERSIDE INDUSTRIAL HEATING PLANT

Experiments were also carried out at the heating plant located at Summerside, Prince Edward Island, Canada. This facility has two identical fluidized bed combustion boilers that can generate a maximum of 18000 kg steam per hour. One of the boilers was used to generate the test data used in this work. The boilers are of two-drum water-tube design utilizing natural circulation. The furnace in each boiler, shown in Figure 11, is divided by a membrane-tube wall into two sections designated as the preferential bed and secondary bed. The internal cross-sectional dimensions are 1.2 x 2.9 m (4' x 9.5') and 1.4 x 2.9 m (4.5' x 9.5') with the secondary bed slightly larger than the preferential. This two-bed system allows for greater flexibility for turn-down. Both beds are surrounded by membrane walls. However, only the secondary bed has immersed heat transfer surface in the form of 18 inclined tubes passing through it. A multicyclone is used to capture particles greater than 40 μm in diameter. The cyclone houses five small-diameter cyclone tubes which operate in parallel. All solids collected in the cylcone and the convective tube bank are recycled to the preferential bed only. The coal, 32 mm x 0, is fed overbed by means of a separate spreader stoker for each bed. The limestone is fed at the opposite end of each bed from the spreader stokers by means of a drop pipe placed at the upper level of the bed. The instrumentation for the flue gas analysis and data acquisition is similar to that installed at the pilot plant at CCRL.

The present data was collected during boiler acceptance tests using the design coal and a series of demonstration trials using four different Maritime coals as alternative fuels. Also included in this report are the data collected during two segments, seven days and three days long respectively, of a 30-day continuous test. This test was carried out early in 1986 for the U.S. Environmental Protection Agency to establish the Ca/S molar ratio required for a minimum sulphur capture of 90% during long-term operation. The Havelock limestone (8 x 20 mesh) was chosen as sorbent. Both the preferential and the secondary beds were in operation during all of the tests discussed here. The chemical analysis data for the coals tested and the operating conditions for these tests are given in Tables 3 and 7.

Figure 12 compares the sulphur retention data calculated from model 1 with that actually achieved. The pilot-scale tests conducted at CCRL suggested that the 18 x 20 mesh Havelock limestone would best represent the in-bed sorbent size for determination of the overall rate of sulphation reaction. The limestone reactivity data employed are the same as those used in Figure 5. The relative high fluidizing velocity employed in these tests also suggested that the sulphur capture was more likely to be underpredicted as mentioned previously, particularly, as there are no tubes in the preferential bed to prevent the bubbles from rapid bubble growth or rise. The assumption of slow bubble regime may, therefore, partly break down for this bed. However, an opposing effect on sulphur retention also existed because of the overbed feeding mode employed. The SO_2 concentration in the bed may be closer to that described by model 3 and the sulphur retention might be expected to be over-predicted by the model used. As a result of these two contradictory influential factors, model 1 agrees well with the experimental data, although the sulphation data show more scatter than that for the tests conducted at the pilot plant at CCRL. The U.S. EPA test probably represents the best experimental data because the unit was operated over a long period and the boilers were operating near optimum performance as a result of major modifications made to the two boilers.

Figure 13 shows typical data of sieve analysis for the four solids streams from test No. 1. The average size of the bed material, defined as the diameter at which the distribution function equals 1/2, was shifted compared with both the coal or the limestone fed, indicating the effect of attrition and breakage. The bed material contains more large particles than the limestone fed suggesting a contribution of the ash produced from the large coal particles. Despite differences in the coal, its size distribution and the Ca/S molar ratio employed, sieve analysis for the bed material of all the 10 tests indicate that the average particle size of the bed material falls between 16 and 20 mesh, i.e., 0.85-1.18 mm, which is close to the effective size of the in-bed sorbent used in calculation for Figures 9 and 10. This suggests that the effective size for the in-bed sorbent in industrial plants may be determined by referring to the average size of the bed material and the results form pilot plant trials.

CONCLUSIONS

Three one-phase analytical models for sulphur retention in fluidized bed combustors operating in small bubble regime have been developed and compared with trials at two pilot plants and an industrial heating plant. The combustor is simplified by a reactor with plug flow or well mixed flow through it. The overall rate of SO_2/CaO reaction of the in-bed sorbent with a wide particle size distribution is assumed to be of first order and is represented by the performance of the sorbent with a narrow size, defined as the effective size for the in-bed sorbent, in a bench-scale fluidized bed reator. The rate constant is expressed as a function of the fraction of sorbent sulphated according to a modified plug pore model. Therefore, the sulphur retention can be related to the operating conditions by means of a single analytical expression for these models.

The plug flow model assuming that the sulphur from coal is released at the base of the bed best describes the SO_2 concentration profiles measured at Point Tupper pilot plant and agrees well with the sulphur retention data achieved in the trials at the pilot plants at CCRL and Queen's University and the tests at Summerside heating plant. This model is recommended for general application. The well mixed flow model tends to underpredict the sulphur retention performance when in-bed coal feeding mode is employed. The model predicts an unreasonably high Ca/S molar ratio for high sulphur retention. The plug flow model assuming a uniform release of the sulphur from coal throughout the bed gives even worse results.

The average size of the sorbent, defined as the diameter at which the distribution function of the sorbent particle size equals 1/2, can be used as the effective size for the in-bed sorbent when the average size is not large. When large particles of limestone are used, 1.55 mm diameter on average for the present work, the effective size is reduced by attrition and breakup of the sorbent in the bed. The effective size for the in-bed sorbent in the Summerside heating plant was found to be the same as that at CCRL when 8 x 20 mesh Havelock limestone was used. The effective size is also close to the average size of the bed material determined by sieve analysis.

The present analysis shows that the volume occupied by the in-bed heat exchange tubes plays no role in the sulphur retention. The effect of the freeboard is also automatically included in the models. The sulphur retention increases primarily with the Ca/S molar ratio employed and also with the operating parameter group q which is a product of gas residence time, solids residence time and the feeding rate of the sulphur from the coal per unit volume of the bed. To ensure simplicity, the models do not take the effects of specific configuration of individual combustor into account. These effects should be considered when the model is applied. Recycle of the cyclone ash increases the fines of the in-bed sorbent and thus decreases its effective size, whereas a bed overflow system reduces the fines in the bed and has the opposite effect. High velocity enhances the attrition and breakup of the sorbent particles and therefore reduces the effective size. If the velocity increases further ($>$3.5 m/s), however, the combustor will not operate in the small bubble regime and the one-phase model will break down.

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Table 1 - Reactor models for sulphur retention in FBC
(see Table 2 for description)

References	A	B	C	D	E
Bethel et al. 1973 (12)	a	b	b&c	c	no
Newby et al. 1980 (13)	a	b	b&c	b	no
Daniel & Finnigan 19890 (14)	a	b	d	d	no
Lee & Georgakis 1981 (15)	b	c	a	a	no
Zheng et al. 1982 (16)	b	a	a	a	no
Fee et al. 1983 (6)	b	a&c	a	a	no
Fee et al. 1984 (11)	b	c	a	a	yes

Table 2 - Description of the models listed in Table 1

A Model for fluidized bed
  a One phase
  b Two phase: bubble phase and emulsion phase

B Gas flow pattern through the bed
  a Well mixed flow
  b Plug flow
  c Plug flow in the bubble phase, mixed flow in the emulsion phase

C Location of sulphur release
  a Uniform release throughout emulsion phase
  b Uniform release throughout the bed
  c Complete release at the base of the bed
  d Release is proportional to char and volatiles combustion

D Kinetics of SO_2/CaO reaction
  a Empirical rate model
  b Data from thermogravimetric analyzer (TGA)
  c Data from differential reactor

E Is SO_2 capture in freeboard considered?

Table 3 - Analytical data for coals

	Evans	Minto	Brogan	Devco	Novaco	Lingan
Proximate analysis:						
Moisture	1.9	0.8	4.9	8.9	6.8	0.5
Volatiles	35.0	33.7	35.5	30.9	32.6	32.5
Fixed carbon	53.4	49.3	50.5	47.5	48.1	48.8
Ash	11.7	17.4	9.0	12.7	12.6	14.4
Sulphur content	6.8	7.4	4.3	4.5	4.1	2.2

Table 4 - Average operating conditions during the EMR sampling program at the Point Tupper unit

	Mean value	Standard deviation
Coal flow (kg/h)	360	60
Limestone flow (kg/h)	60	6.0
Air flow (kg/h)	2825	56
Flue gas O_2 (vol %)	3.03	0.14
Bed temperature (°C)	851	3.9
Bed height (mm)	1275	14
Recycle rate (kg/h)	513	15

Table 5-Operating conditions for trials at Queen's University

Trial	Coal size mm	Coal rate kg/h	Stone rate kg/h	Bed temp. °C	Vel. m/s	Ca/S	Solids resid. time h	Recycle fraction
Evans coal, 0.15-0.99 mm (16x100 mesh) Havelock limestone								
831115	16.7	46.4	39.0	869	2.1	4.14	2.1	0.0
831122	16.7	43.9	15.3	877	2.1	1.72	4.4	0.0
831124	16.7	48.3	17.7	865	2.2	1.80	4.6	0.555
840313	16.7	24.2	13.6	858	1.5	2.77	7.9	1.0
840314	16.7	22.5	13.2	915	1.6	2.89	6.3	0.0
840320	16.7	20.7	7.4	879	1.5	1.75	9.2	0.0
840207	8.8	32.7	9.7	897	2.1	1.43	6.8	0.0
840215	8.8	41.7	20.3	908	2.3	2.35	3.9	0.555
840216	8.8	35.7	20.0	837	2.2	2.70	3.5	0.0
840124	3.3	26.6	14.6	831	1.5	2.83	5.4	0.963
840126	3.3	26.8	13.8	874	1.6	2.67	5.2	0.0
Minto coal, 0.15-0.99 mm (16x100 mesh) Havelock limestone								
830322	19.3	31.1	15.1	840	2.1	2.14	3.5	0.0
830324	19.3	34.4	14.6	855	2.1	1.87	3.9	0.70
830329	19.3	38.2	31.0	855	2.3	3.73	2.2	0.59
830330	19.3	37.4	30.3	850	2.0	3.57	2.4	0.0
Minto coal, 0.44-1.5 mm (11x34 mesh) Havelock limestone								
821116	19.3	29.3	8.4	850	1.6	1.25	6.7	1.0
830112	10.7	31.0	11.4	820	1.7	1.60	5.1	0.963
830124	10.7	32.8	15.8	820	1.9	2.10	4.3	0.905
830126	10.7	30.3	28.0	805	1.8	4.25	3.0	0.836
830127	10.7	30.3	18.5	835	1.8	2.66	4.1	0.875
830201	6.6	33.3	17.5	825	1.9	2.24	4.1	0.919
830202	6.6	33.1	28.0	840	1.9	3.60	2.8	0.906
830215	6.6	41.0	21.6	860	2.8	2.24	2.8	0.60
830216	6.6	40.9	28.0	865	2.8	2.92	2.4	0.52
830217	6.6	39.8	27.6	840	2.7	2.95	2.3	0.0

Table 6-Operating conditions for trials at CCRL using Brogan coal and 0.85-2.36 mm (8x 20 mesh) Havelock limestone

Test No.	Coal rate kg/h	Bed temp. °C	Vel. m/s	Ca/S	Mass of B.M. kg	Moles of Ca per kg B.M. 10^{-2} kmole/kg	% air used for recycle
BR01	40.81	849	2.11	2.88	155.0	0.4139	0
BR02	38.05	848	2.14	2.95	165.0	0.5240	8.2
BR03	23.56	857	1.33	2.97	143.0	0.4582	0
BR04	22.92	854	1.31	2.97	129.5	0.2948	5.9
BR05	37.82	847	2.17	2.72	159.0	0.5900	7.5
BR06	53.72	845	3.08	2.94	168.0	0.7494	14.9
BR07	53.40	850	3.06	2.34	180.0	0.8183	15.0
BR08	21.48	847	1.27	2.63	154.0	0.4765	8.3
BR09	20.96	853	1.26	3.52	146.0	0.3481	0
BR10	52.40	851	2.86	2.94	197.0	0.8233	0
BR17	33.03	848	2.02	2.39	288.9	0.6339	9.7
BR18	20.86	858	1.21	2.77	290.8	0.9767	7.3
BR19	34.44	898	2.12	2.64	140.0	0.4212	11.6
BR21	17.01	889	1.31	3.44	143.0	0.3507	8.7
BR22	49.32	896	3.07	2.60	160.4	0.7799	19.4

Table 7- Operating conditions for trials at Summerside heating plant

Test No.	Coal Name	Coal S %	Rate kg/h	Boiler load %	Bed temp. °C	Vel. m/s	Ca/S ratio	Solids resid. time h
01	Devco	4.40	1752	100	861	3.52	2.06	11.26
02	Devco	4.60	1177	65	845	2.00	3.04	13.74
03	Brogan	4.81	1686	100	844	3.55	2.53	11.53
04	Brogan	5.18	1087	65	863	2.40	2.61	16.83
05	Evans	6.12	2030	100	863	4.46	3.06	6.91
06	Evans	5.56	1452	65	830	3.44	2.62	10.99
07	Minto	7.38	1698	100	850	3.44	2.39	6.62
08	Novaco	4.63	1774	100	848	3.03	1.98	9.30
*09	Evans	5.90	1405	73	840	2.95	1.91	5.29
*10	Evans	5.91	1338	73	833	2.57	1.35	5.65

* Segment of the U.S. EPA 30-day test.

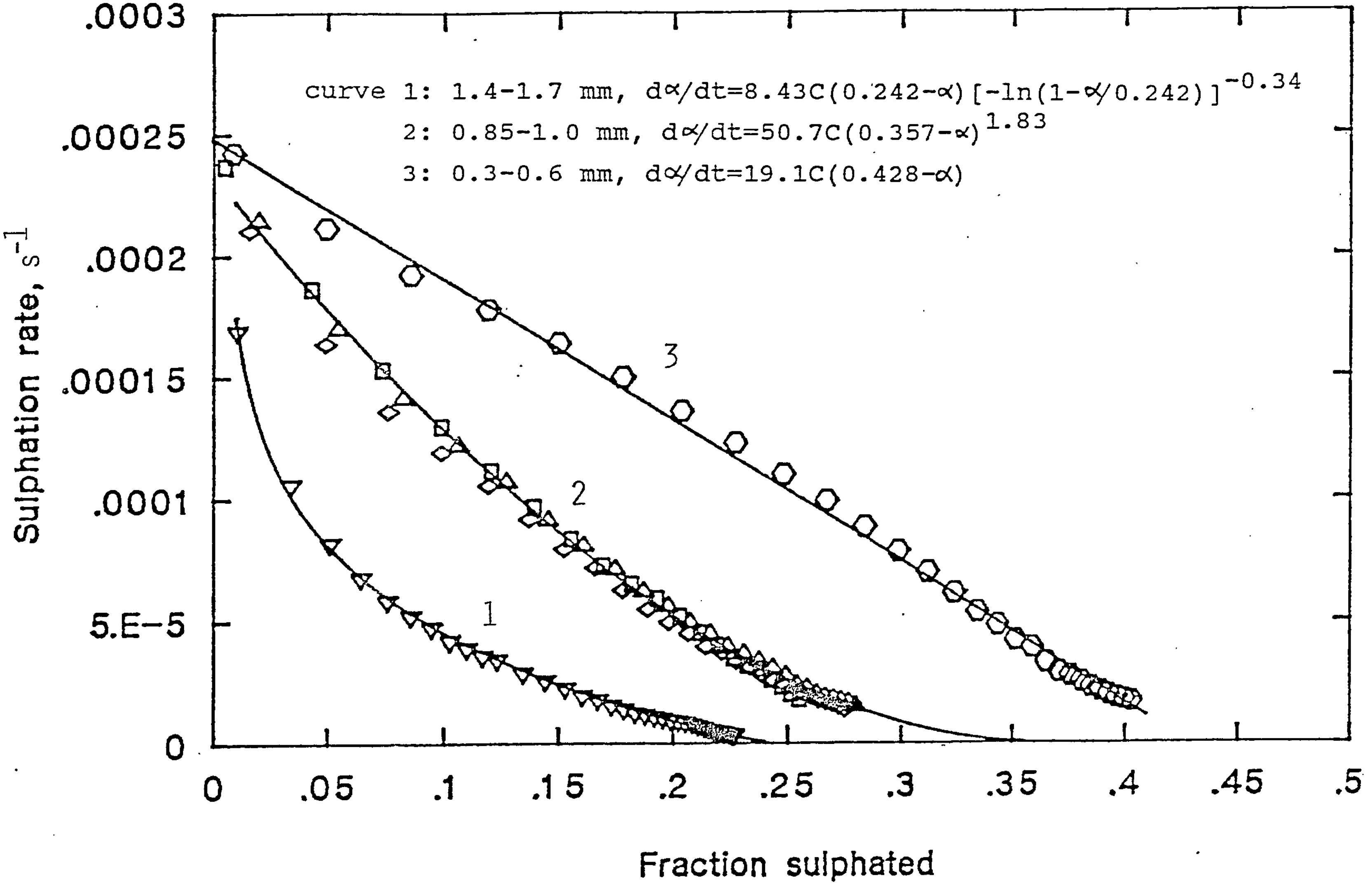


Fig. 1 Sulphation rate for Havelock limestone of different sizes

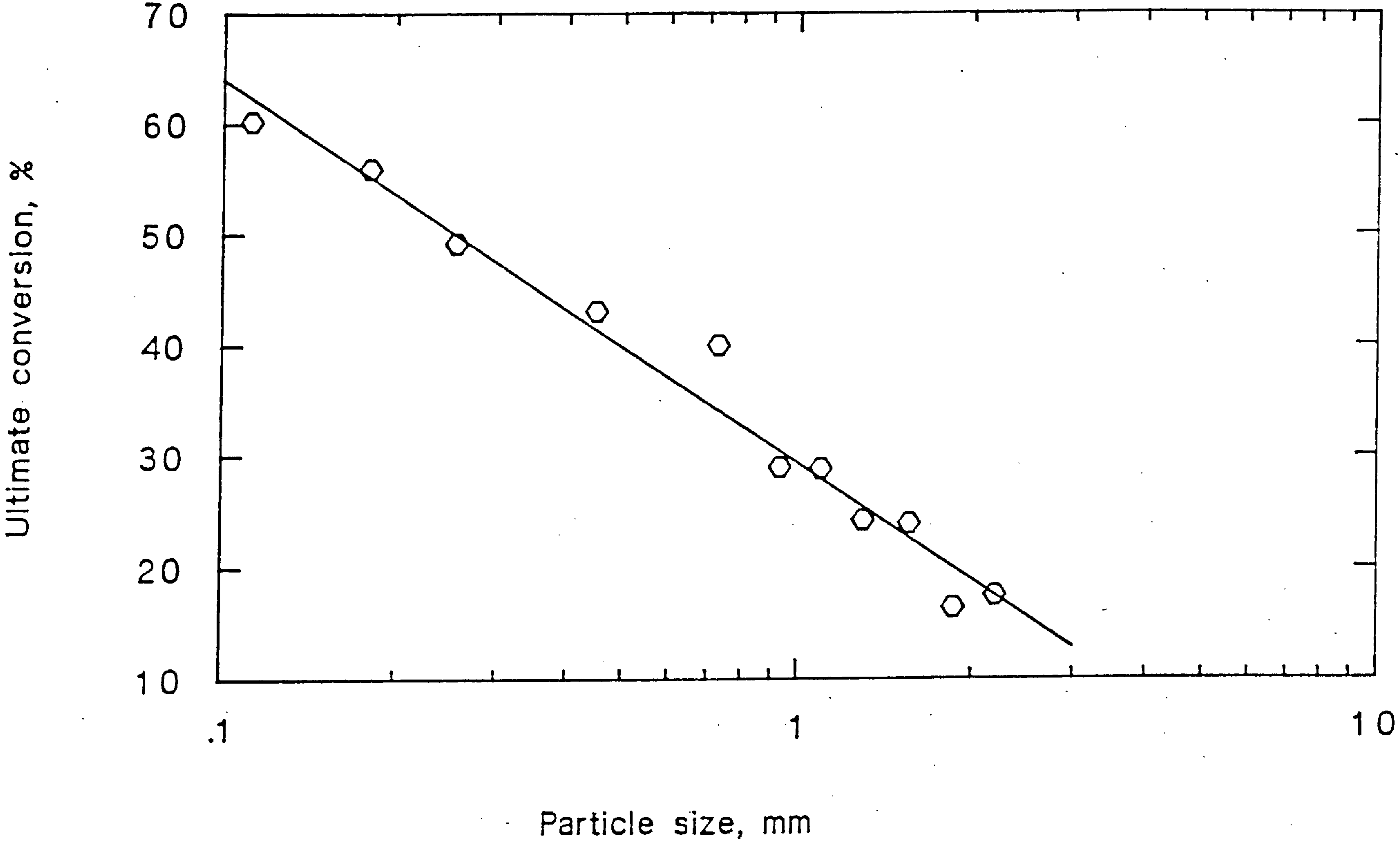


Fig. 2 Ultimate conversion of Havelock limestone versus particle size

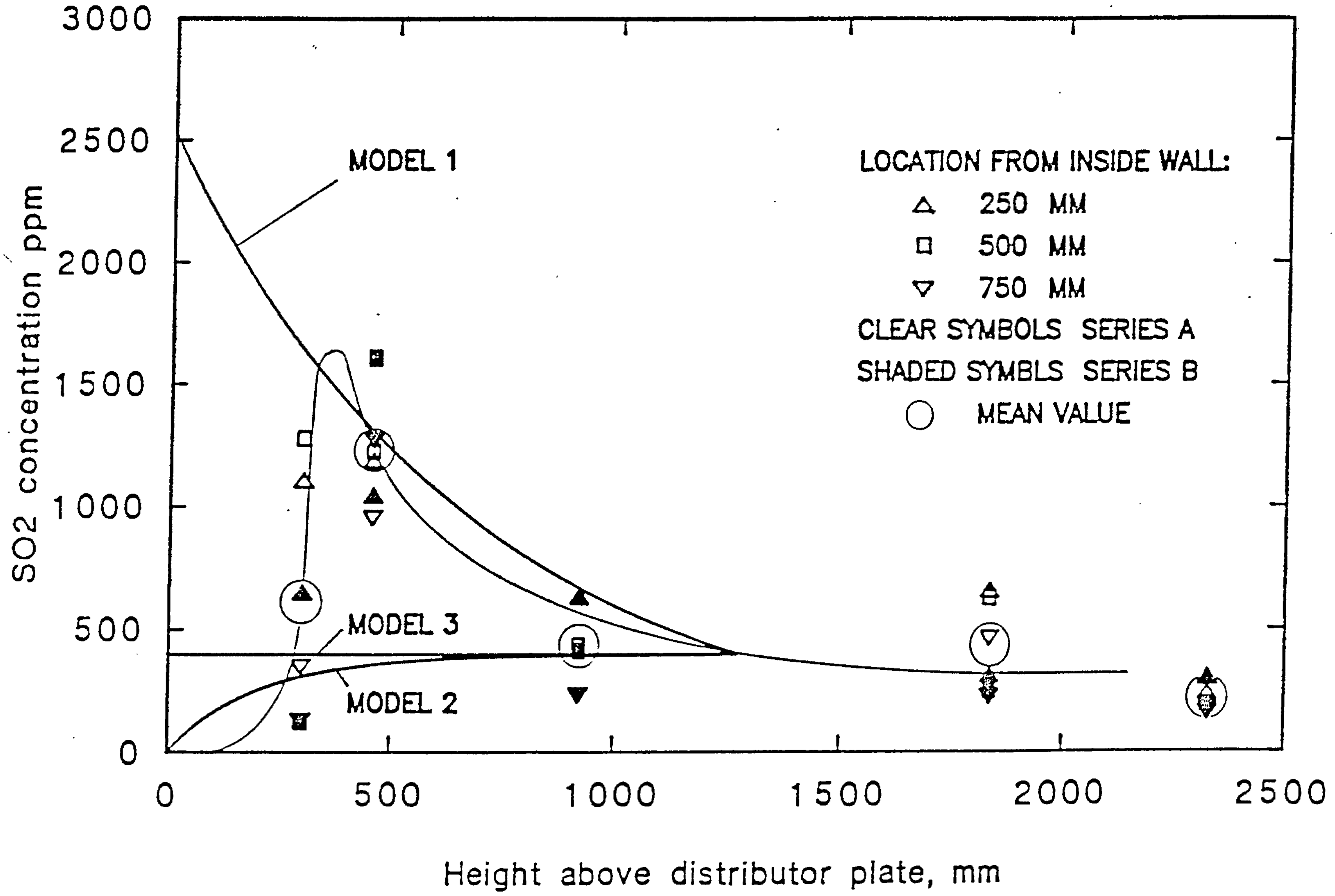
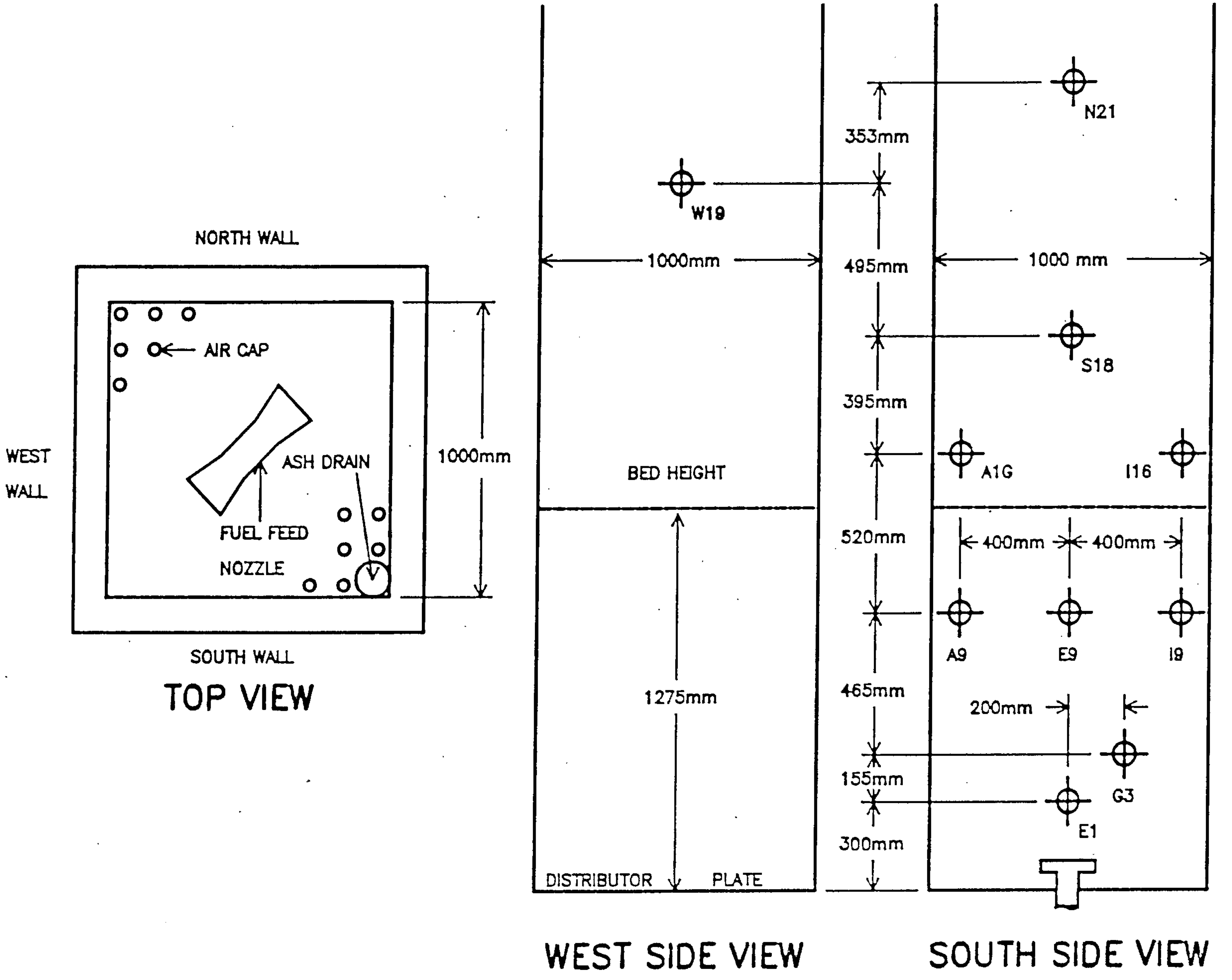


Fig. 3 COMPARISON WITH MEASURED SULPHUR DIOXIDE CONCENTRATIONS IN THE POINT TUPPER UNIT



TOP VIEW

WEST SIDE VIEW

SOUTH SIDE VIEW

Fig. 4 Position and LABELLING of sampling ports at the Point Tupper unit

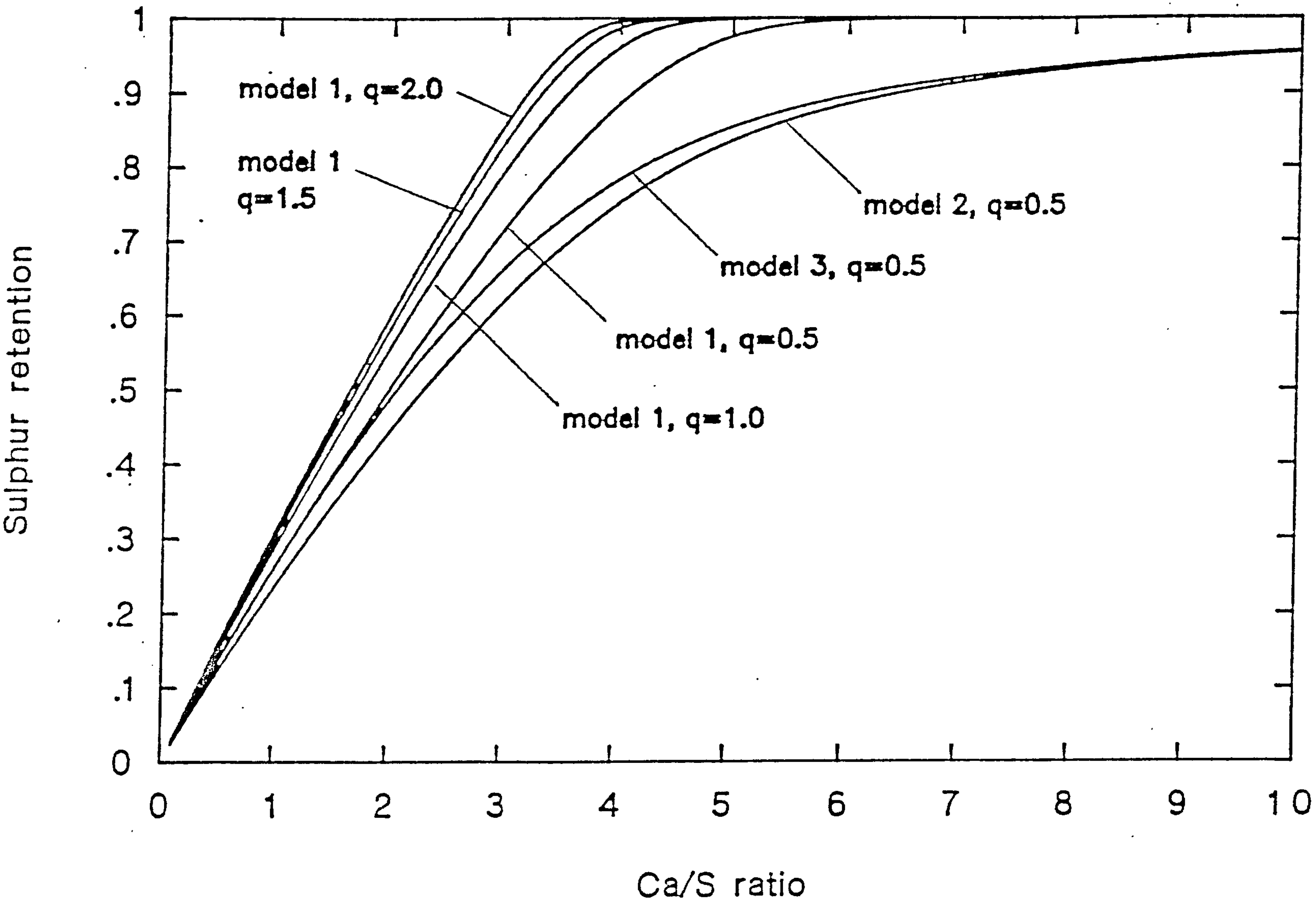


Fig. 5 Variation of sulphur retention with calcium to sulphur molar ratio

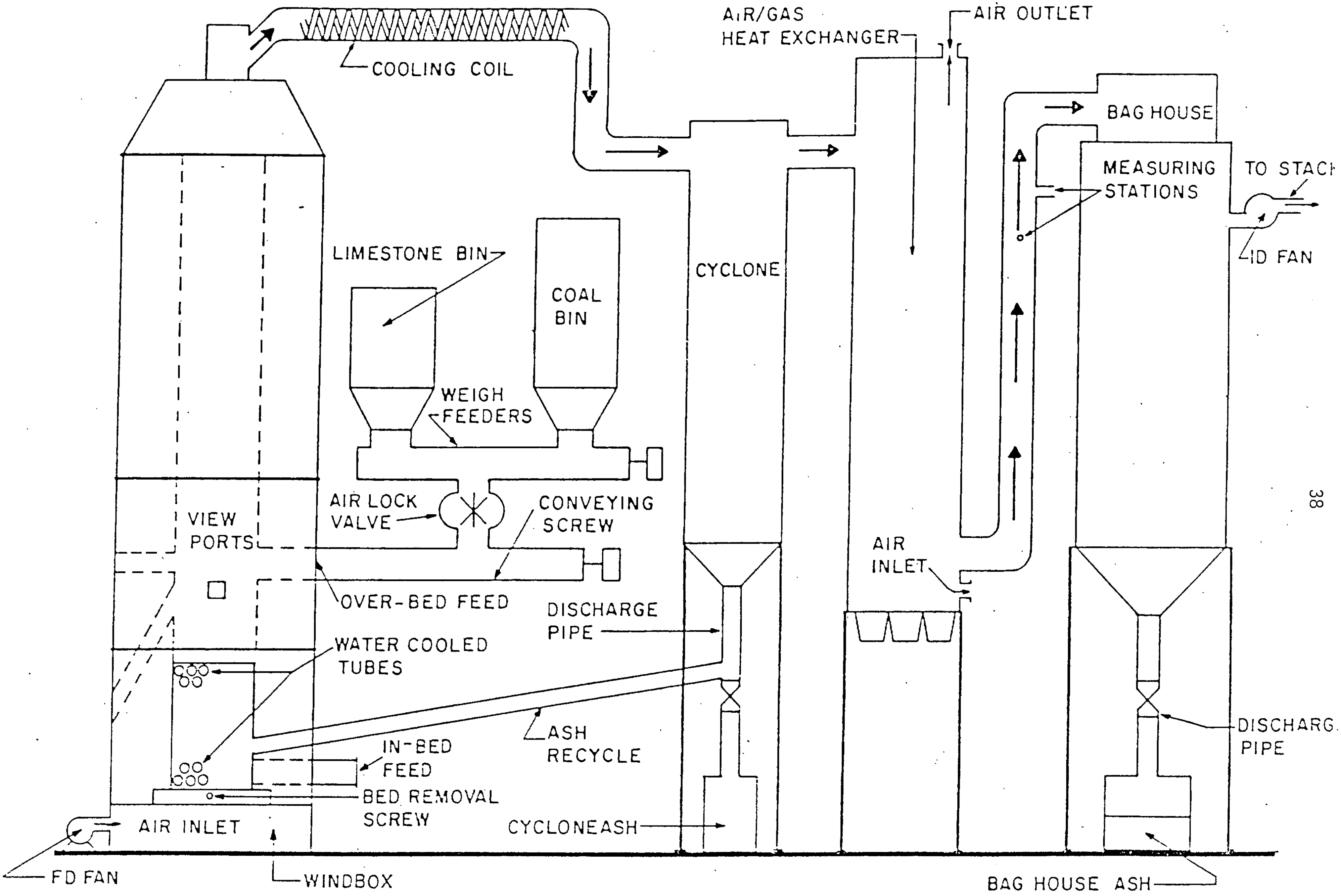


Fig. 6 SCHEMATIC OF THE CCRL BUBBLING FLUIDIZED BED COMBUSTOR

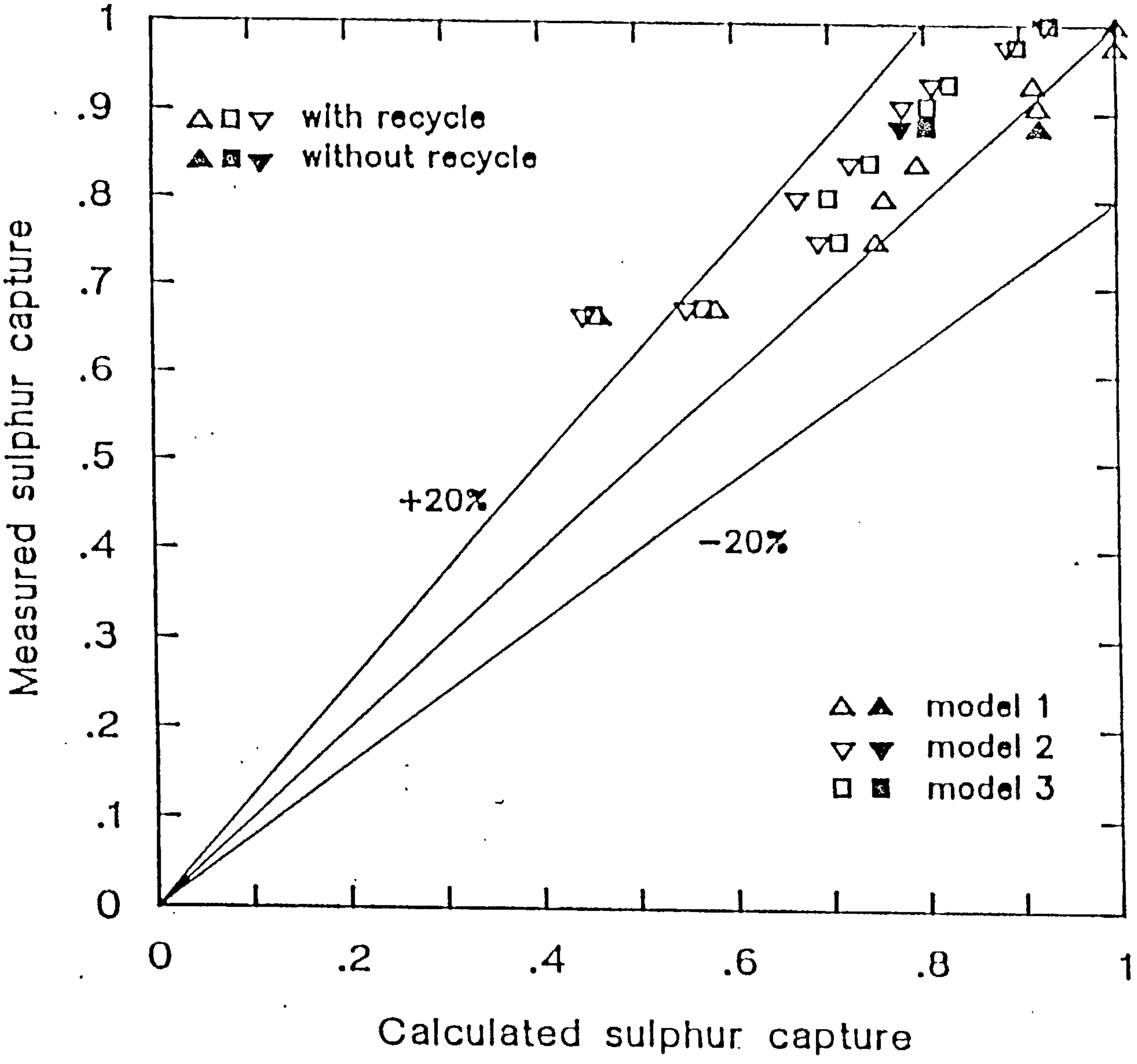


Fig. 7 Comparison of calculated and measured sulphur capture data with Minto coal and 11 x 34 mesh Havelock limestone

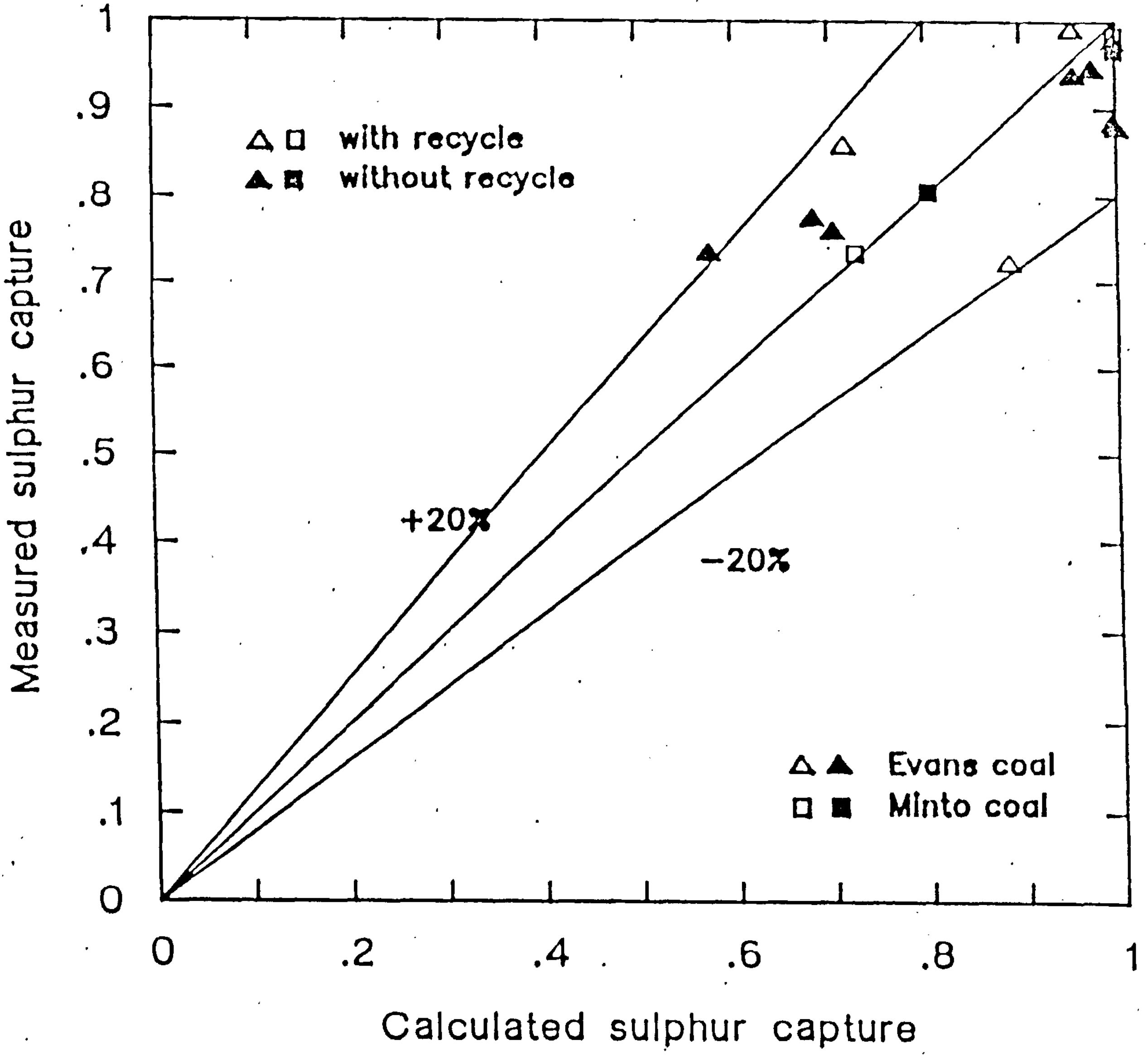


Fig. 8 Comparison of calculated (from model 1) and measured sulphur capture data with 16 x 100 mesh Havelock limestone

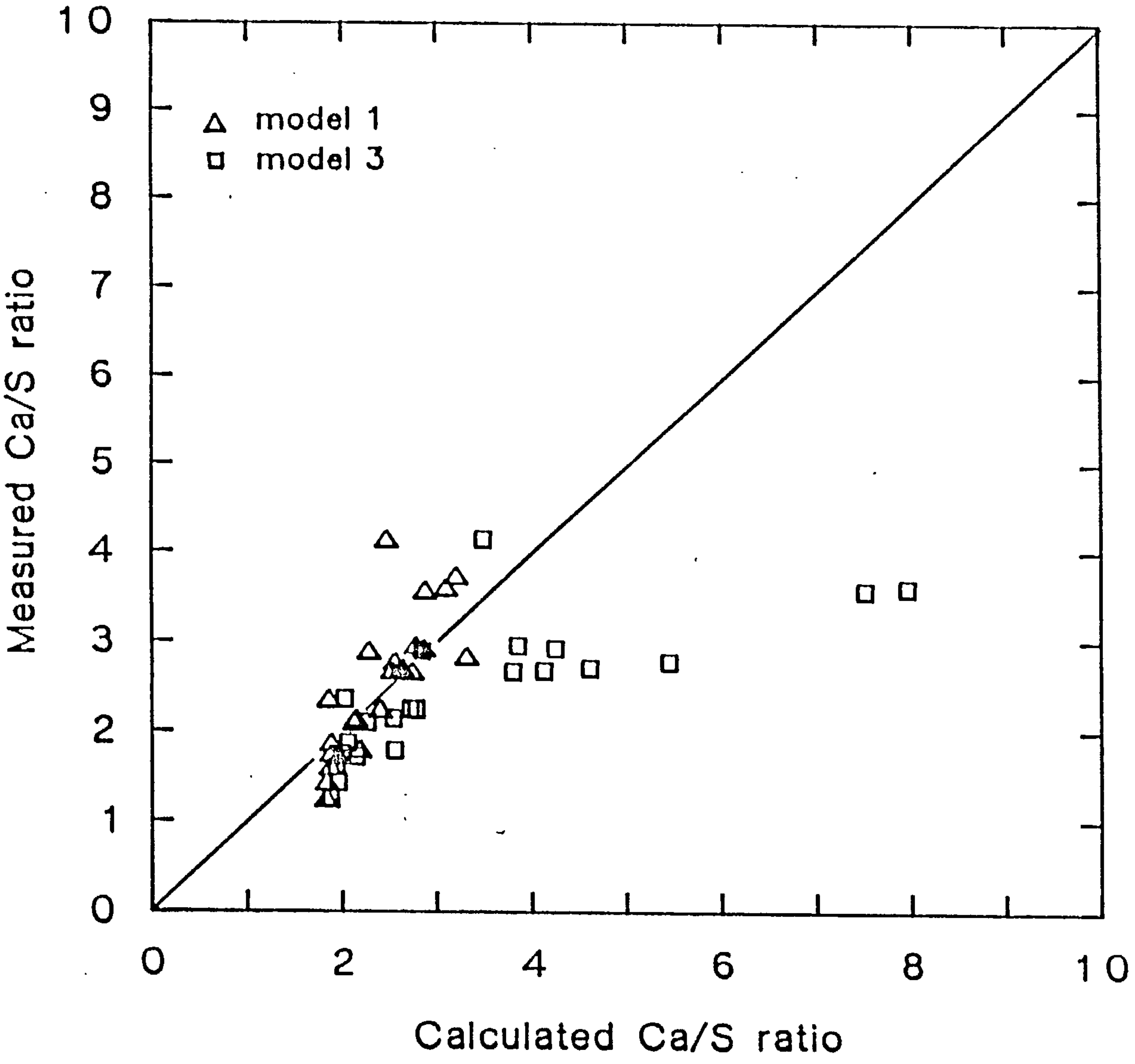


Fig. 9 Comparison of calculated and actual calcium to sulphur molar ratio with 11x34 mesh and 16X100 mesh Havelock limestone

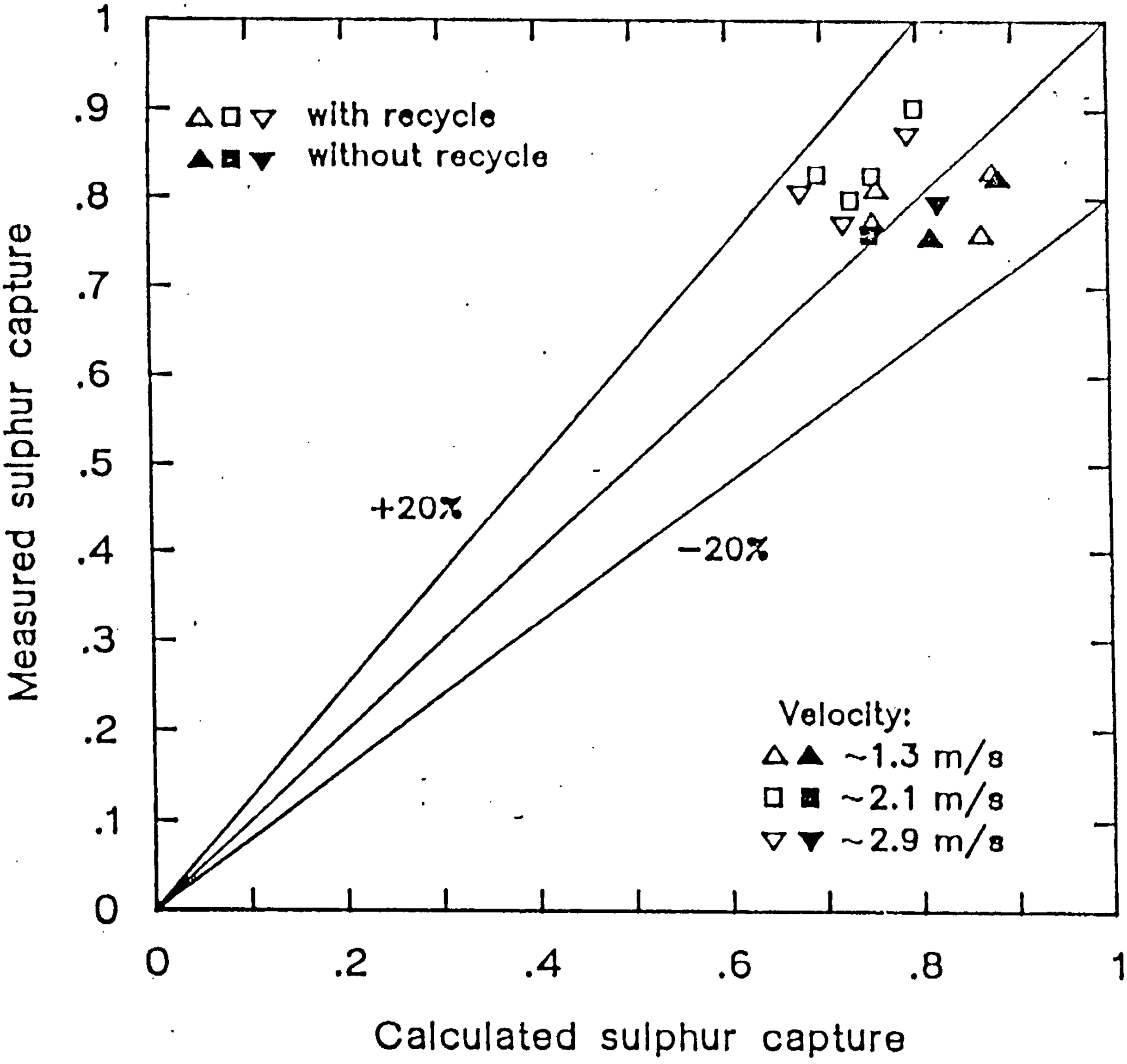


Fig. 10 Comparison of calculated (from model 1)and measured sulphur capture data with Brogan coal and 8x20 mesh Havelock limestone

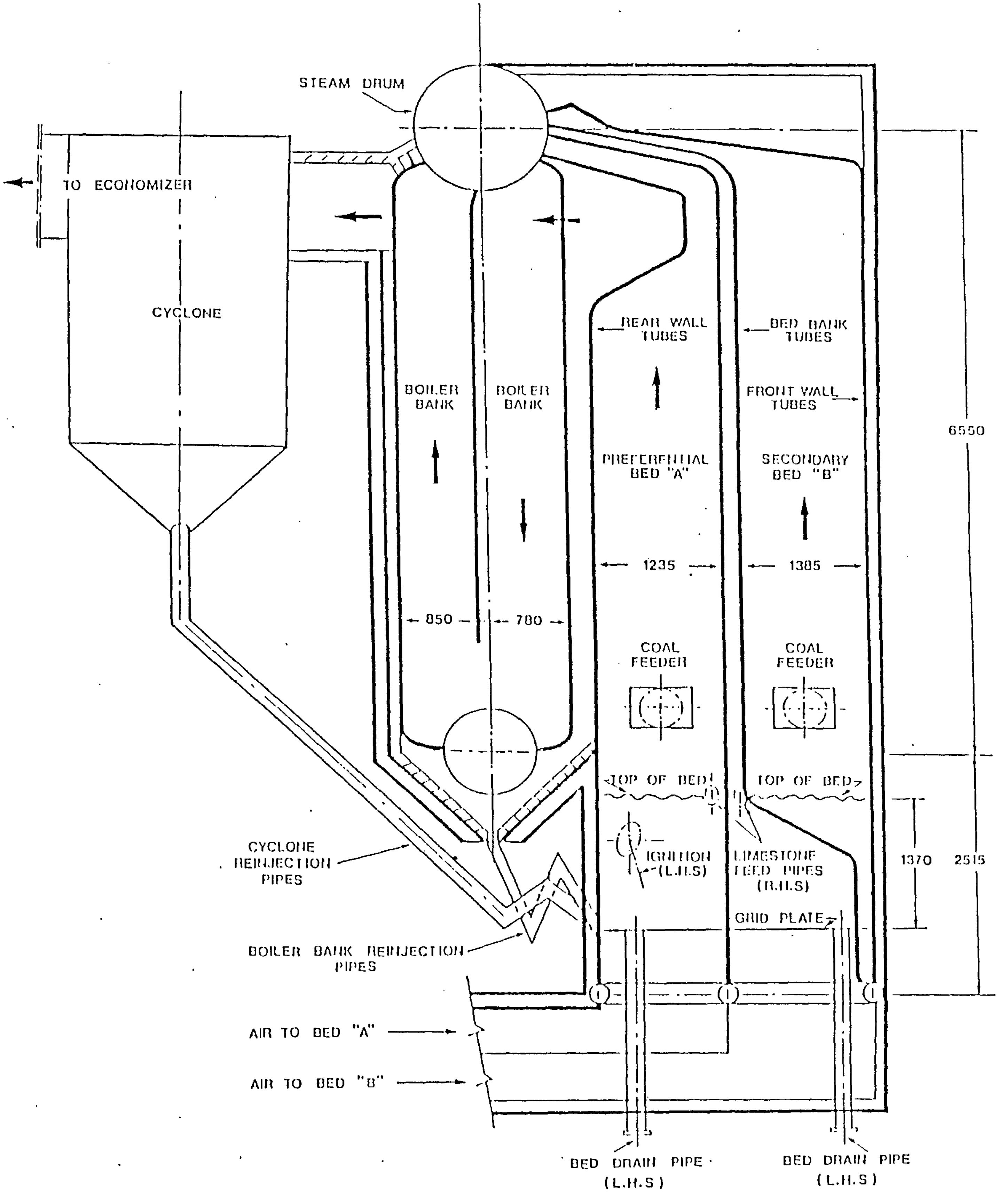


Fig. 11 Section view of a fluidized bed boiler at Summerside

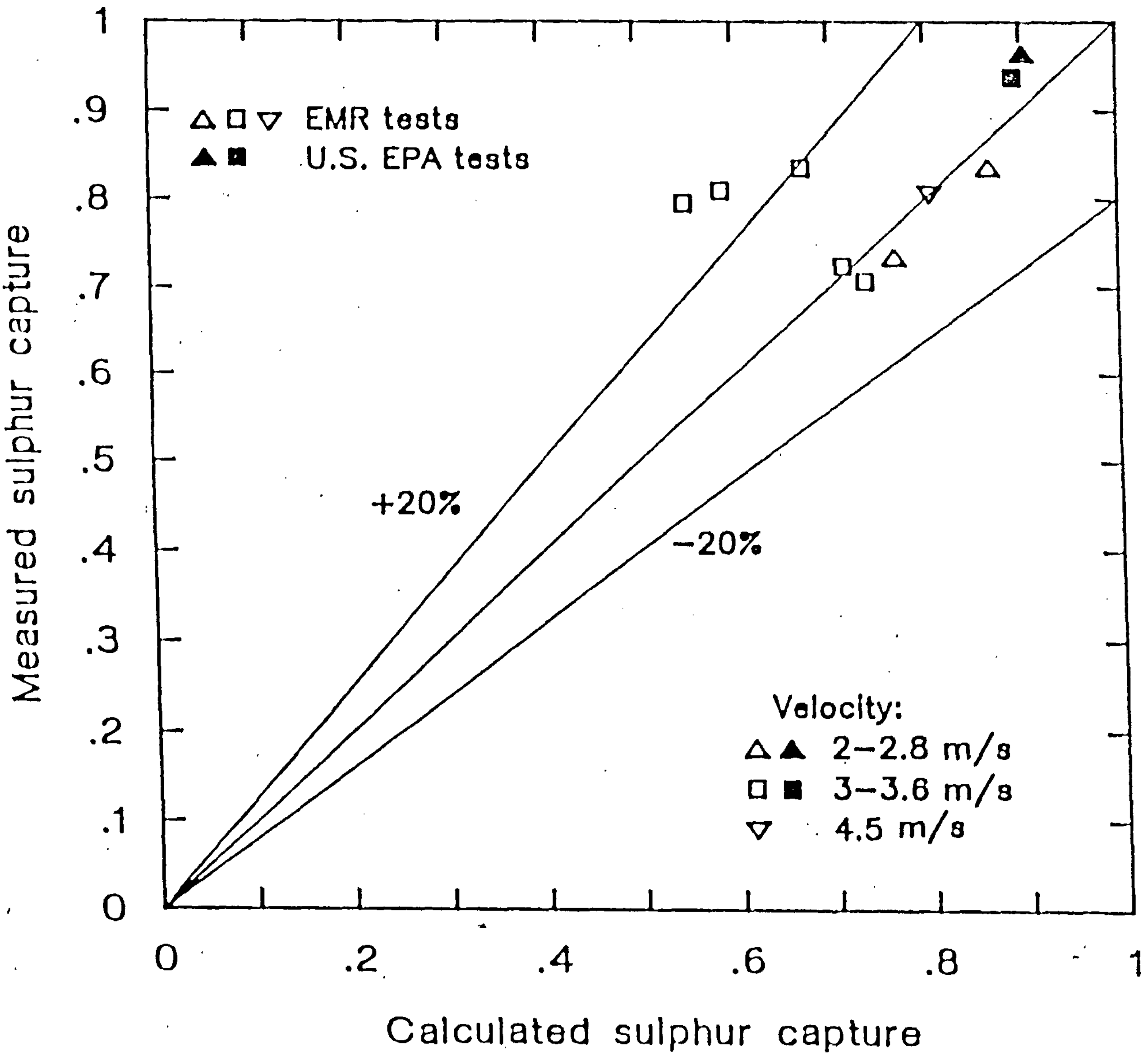


Fig. 12 Comparison of calculated (from model 1) and measured sulphur capture data at Summerside heating plant

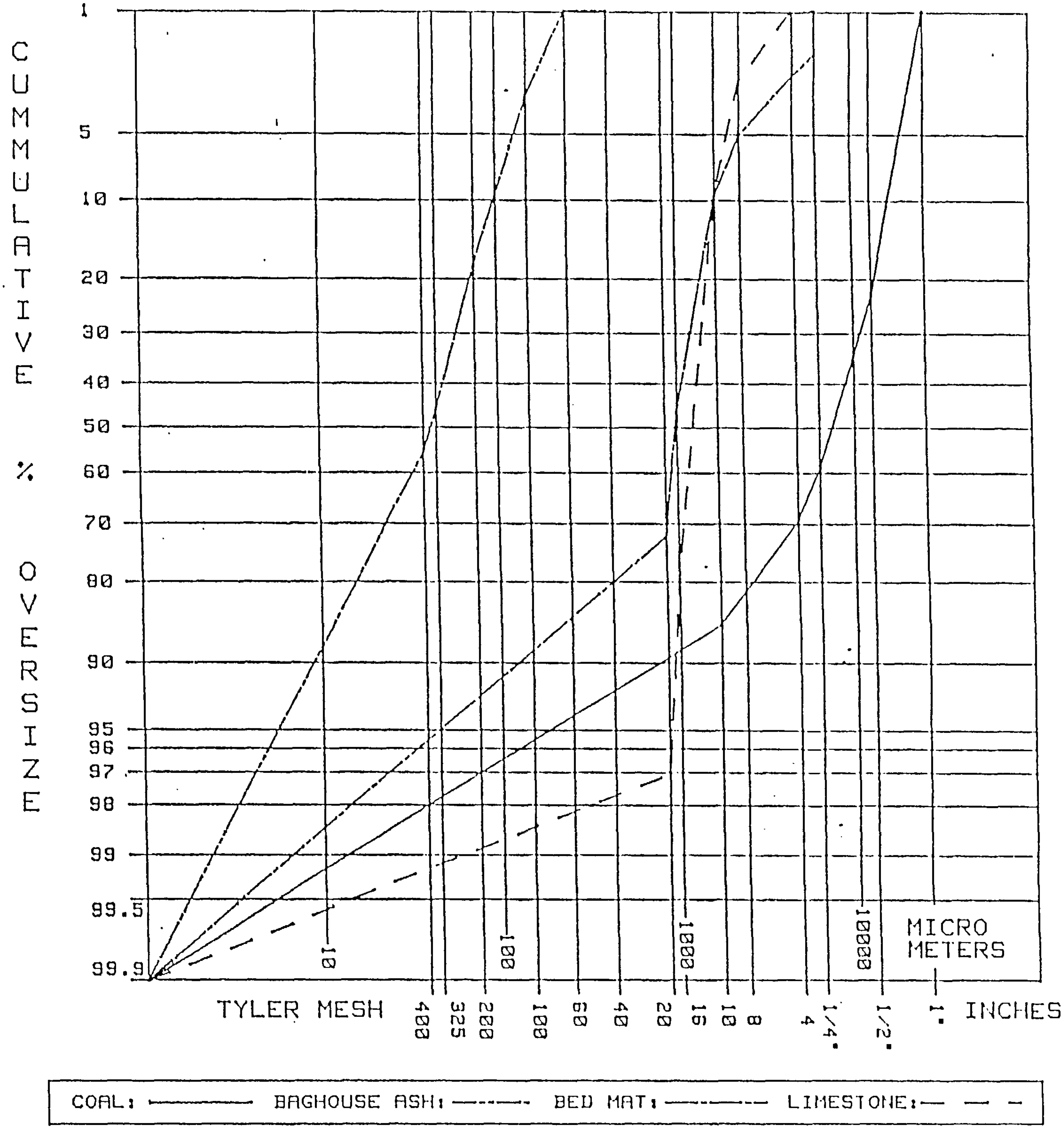


Fig. 13 Sieve analysis of solids streams from Summerside heating plant in test No. 1

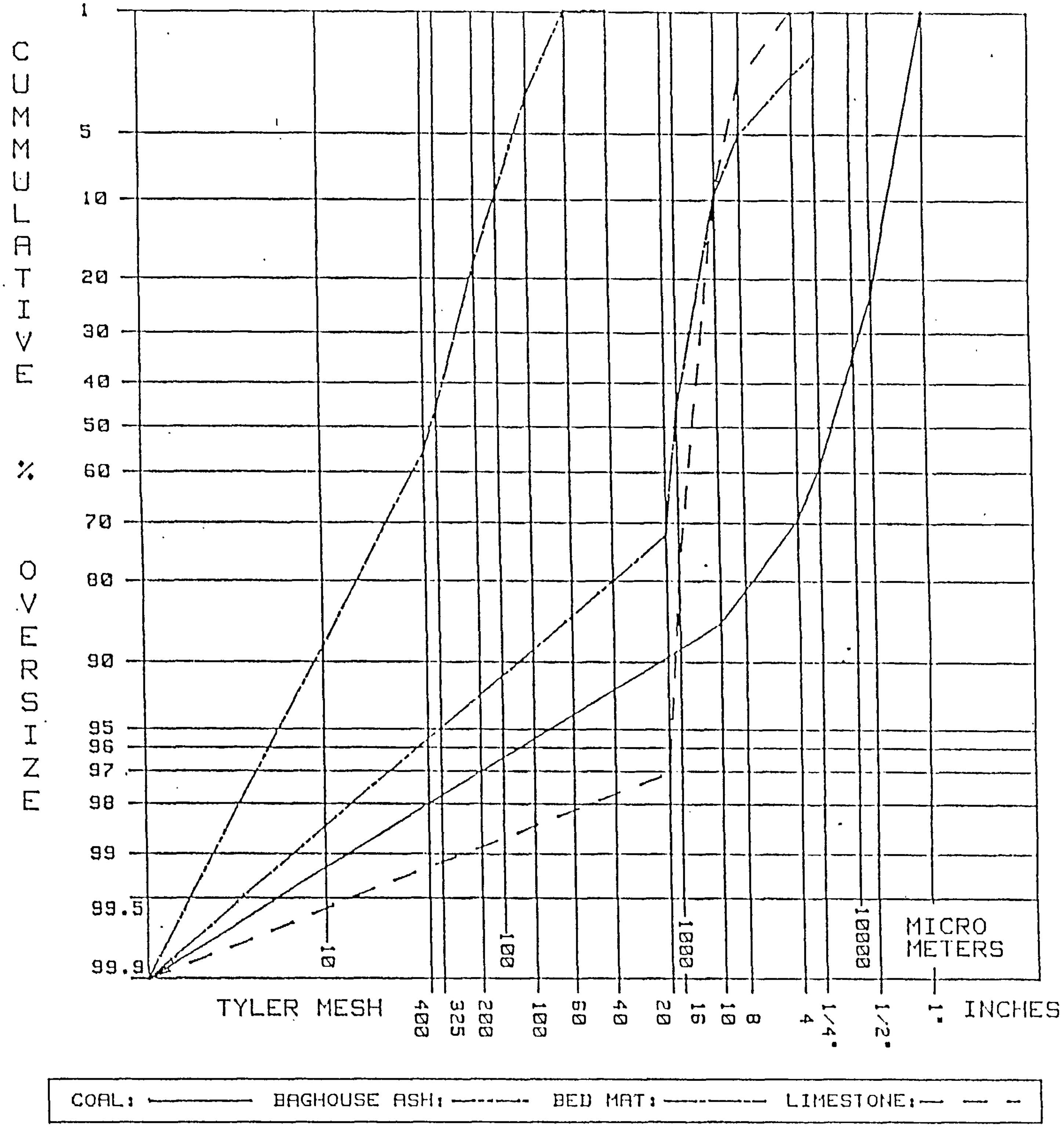


Fig. 13 Sieve analysis of solids streams from Summerside heating plant in test No. 1