

Development of Linear Model for Predicting the Surface Area of Sorbent Prepared from Coal Fly Ash : Statistical Design Approach

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(Received : 31 January 2004 – Accepted : 15 March 2004)

Abstract: Active sorbent for flue gas desulphurization was prepared from coal fly ash, calcium oxide (CaO) and calcium sulfate (CaSO₄) using hydro-thermal process. The effect of various sorbent preparation variables; hydration period (A), amount of fly ash (B), amount of CaSO₄ (C) and drying temperature (D) towards the BET (Brunauer-Emmett-Teller) specific surface area of the resulting sorbent were studied using statistical design of experiment. The surface area obtained ranges from 12.9 to 92.7 m²/g. Based on the probability plot for

the effect of various variables, it was concluded that all the individual variables studied except for variable D exerted significant influence on the surface area of the sorbent. Apart from that, the interaction between variable A and B are also significant. From this finding, a linear mathematical model that correlates the sorbent preparation variables to the BET surface area of the sorbent was developed. Analysis of variance (ANOVA) showed that the model was significant at a confidence level of 99%. The absolute average percentage deviation of the results simulated by the model as compared to experimental results is less than 10%.

Keywords: Coal ash, Linear model, Sorbent, Statistical design, Surface area.

Introduction

Environmental regulations all over the world are becoming more restrictive concerning the release of atmospheric pollutants associated with the flue gases of combustion systems, particularly the emission of sulphur dioxide (SO₂). SO₂ is the primary cause of acid rain, which damages buildings, vegetation and water ground cycles. SO₂ also causes the formation of secondary particles in the atmosphere that impairs visibility. Inhaling SO₂ into the human respiratory tract can also pose a hazard towards human health as it could cause lung and throat diseases. Table 1 shows typical flue gas composition from a coal-fired power plant of 600 MW capacity burning coal

containing 1.2% sulphur [1]. From Table 1, it can be seen that the concentration of SO₂ is at 350 ppm, whereas most environmental regulations throughout the world only permits about 50 to 100 ppm. Therefore, flue gas from the power plant has to be treated with appropriate yet affordable desulphurization technology before it can be emitted to the environment.

Table 1. Typical flue gas composition.

Components	Concentration
SO ₂	350 ppm
NO _x	125 ppm
O ₂	5.2 %
CO ₂	13 %
H ₂ O	7.8 %
N ₂	Balance

One process currently under active development is the utilization of coal fly ash in the preparation of sorbents for dry flue gas desulphurization (FGD). Various studies have shown that when silica (SiO₂, eluted from coal fly ash) is mixed with calcium hydroxide, Ca(OH)₂ or calcium oxide, CaO, in a hydration process, sorbent with high SO₂ capture efficiency could be attained [2-6]. This is due to the pozzolanic reaction between Ca(OH)₂/CaO and SiO₂ that produce highly hydrated products that has a high surface area. It was also reported in the

literature that the addition of calcium sulphate, CaSO_4 into the preparation mixture can further promote the formation of the hydrated products [7]. The use of coal fly ash as the sorbent is attractive both economically and environmentally, as it is the most voluminous by-product from coal-fired power plants [8]. The sorbent, after reaction with SO_2 is converted into an eco-friendly product which can be disposed off easily due to its multifunctional usages. This includes its application as fertilizer, coagulating agent, deodorizer for refrigerators, shoes and even pet litter.

The major product of the pozzolanic reactions is believed to be hydrated calcium alumino silicates ($\text{Ca}_x\text{Si}_y\text{Al}_w\text{O}_z \cdot n\text{H}_2\text{O}$) with different values of n, w, x, y and z , depending on the methods of sorbent preparation as well as the composition of coal fly ash used and sorbent preparation variables. Different composition of the sorbent will lead to sorbent with different structural properties. Many researchers have then reported on the effects of different structural properties of the sorbent such as BET surface area on the SO_2 capture efficiency of the sorbent. Jozewicz and Chang, 1989 [9] reported that high sorbent surface area is one of the necessary factors that must be found in a sorbent for it to be able to react with SO_2 under the conditions encountered in dry-type FDG processes. In the paper, the importance of higher sorbent surface area on the SO_2 capture was highlighted. Fernandez *et al.*, 1997 [8] reported that the SO_2 capture activity of these sorbents generally increased with higher BET surface area of the sorbent.

In 2003, Lin, *et al.*, [10] reported that the utilization of Ca in the sorbent increased linearly with increasing BET surface area of the sorbent.

As reactivity seems to correlate with the surface area of the sorbent, several studies [3,11-14] have reported on the correlation between the sorbent preparation variables to the surface area of the resulting sorbent. However, most of the sorbents reported in the literature are prepared from Ca(OH)_2 [7]. Recently, Ishizuka *et al.*, 2000 [15] reported that the activity of sorbent further increases when CaO is used instead of Ca(OH)_2 as the raw material for preparing the sorbent. Thus, this work intends to investigate the correlation between the sorbent preparation variables to the resulting sorbent surface area for sorbent prepared from coal fly ash, CaSO_4 and CaO instead of Ca(OH)_2 . The effect of four sorbent preparation variables ; hydration period (A), amount of fly ash (B), amount of CaSO_4 (C) and drying temperature (D) towards the BET surface area of the sorbent were studied. Statistical design of experiment based on full factorial design of 2^4 was used in the present study to establish the significance of each independent variable and interaction between variables on the surface area of the sorbent. Apart from that, a linear mathematical model that correlates the significant variables towards the BET specific surface area of the sorbent was also developed. The validity of the model was verified by comparing simulated with experimental results and analysis of variance (ANOVA).

Besides that, XRD (X-Ray Diffraction) was used to analyze the various phases present in the sorbent.

Materials and Methods

1. Sorbent Preparation.

The sorbents were prepared from calcium oxide (CaO), coal fly ash and calcium sulphate (CaSO₄). The CaO (laboratory grade) and CaSO₄ (reagent grade) used were supplied from BDH Laboratory Supplies, England. The coal fly ash was supplied by Kapar Power Plant, Malaysia of Tenaga Nasional Berhad with the following composition : SiO₂, 60% ; Al₂O₃, 20% ; Fe₂O₃, 4.7% ; CaO, 3.0% ; K₂O, 1.1% ; MgO, 1.0% ; C, 7.5% ; others, 2.4% and ignition loss, 0.3%.

The sorbents were prepared using the water hydration method. 5g of CaO was added to 100 ml of water at 70°C. After that, 5 or 10g of coal fly ash and 3 or 6g of CaSO₄ were added simultaneously into the slurry. The slurry was then heated under reflux at 98°C for 8 or 16 hrs. hydration time. Upon completion of the hydration period, the slurry is then filtered and dried in an oven at 200 or 500°C.

2. Physical and Chemical Analysis.

The sorbents were analyzed for their BET specific surface area (calculated using the BET standard method) using Autosorb 1C Quantachrome. XRD spectrum were recorded on a Philips

PW 1820 system for the powdered samples with Cu-K α radiation in the range of diffraction angle (2θ) 5°-90° at a sweep rate of 3 deg/min.

3. Statistical experimental design.

A 2 level full factorial design of experiments was used to evaluate the significance of four sorbent preparation variables on the BET specific surface area of the sorbent. Table 2 lists the range and levels of the four independent variables studied in terms of coded and actual values.

Table 2. Experimental variables studied.

Experimental variables	Units	Coding	Range and levels	
			Low (-1)	High (1)
Hydration period	hrs.	A	8	16
Amount of fly ash	gram	B	5	10
Amount of CaSO ₄	gram	C	3	6
Drying temperature	°C	D	200	500

The relation between coded independent variable A with the actual values is given as in Eq. (1)

$$A = \frac{(A_i - A_0)}{\Delta A} \quad (1)$$

where A is the coded value of independent variable A and A_i , A_0 , ΔA are the actual value, actual value at the centre point and the step change of variable A respectively. The relation between coded independent variables B , C and D with the actual values can also be obtained from Eq. (1) similarly. Since there are four variables studied, 16 (2^4) experimental runs with 5 replicates at the central points are required. The complete design matrix and results are shown in Table 3. Note that a particular treatment combination is always represented by lowercase letters and a high level of any variables in the treatment combination is denoted by the corresponding lowercase letter, while the low level of any variables in the treatment combination is denoted by the absence of the corresponding letter. For example, treatment combination c represents the following hydration conditions; variables A , B and D at low level and variable C at high level. All the experimental runs were conducted in randomized order to guard against systematic bias.

Table 3. Experimental design matrix and results.

Treatment combinations	Experimental variables				BET surface area (m ² /g)
	A	B	C	D	
(l)*	-1	-1	-1	-1	28.4
a	1	-1	-1	-1	43.2
b	-1	1	-1	-1	26.4
ab	1	1	-1	-1	58.3
c	-1	-1	1	-1	33.7
ac	1	-1	1	-1	57.7
bc	-1	1	1	-1	39.5
abc	1	1	1	-1	92.7
d	-1	-1	-1	1	24.3
ad	1	-1	-1	1	39.8
bd	-1	1	-1	1	12.9
abd	1	1	-1	1	65.3
cd	-1	-1	1	1	28.9
acd	1	-1	1	1	42.1
bcd	-1	1	1	1	30.2
abcd	1	1	1	1	90.8
S1	12	7.5	4.5	350	59.9
S2	12	7.5	4.5	350	54.3
S3	12	7.5	4.5	350	56.2
S4	12	7.5	4.5	350	61.0
S5	12	7.5	4.5	350	58.2

*(l)- All the variables are at low level

In order to evaluate the significance of the studied variables on the surface area of the sorbent, the method of plotting the effect of a variable on normal probability paper was used [16]. From the probability plot, effects that are negligible are normally distributed, with mean zero and variance σ^2 , will fall along a straight line on this plot, while significant effects will have nonzero means and will not lie along the straight line. The average effect of a variable is defined as the change in response (BET surface area) produced by a change in the level of that variable, averaged over the levels of the other variables.

Results and Discussion

BET surface area for coal fly ash, CaO, CaSO₄ and sorbent obtained under treatment combination abc (maximum value of surface area) are given in Table 4. The significant increase in the BET surface area of the hydrated coal fly ash sorbent as compared to the raw materials suggests that the mixing of the above-shown reagents resulted in the formation of certain compounds with a relatively higher surface area. The higher surface area of the solid sorbent is an important parameter in the solid-gas adsorption (between SO₂ and sorbent) process as it may be one of the principal factors that determine the overall reactivity.

Table 4. BET surface area of raw materials and hydrated sorbent.

Material	BET surface area, m ² /g
Coal fly ash	1.46
CaO	5.62
CaSO ₄	4.89
Prepared sorbent	90.8

XRD analysis was used to determine and analyze the various phases present in the sorbent. Figure 1 shows the XRD spectrum for the sorbent prepared under treatment combination abc. From the diffraction spectrum, it can be deduced that there are 3 main phases present in the sorbent which are, calcium alumino silicate hydrate ($\text{Ca}_2(\text{Si}_9\text{Al}_3)\text{O}_{24}\cdot 8\text{H}_2\text{O}$), calcium carbonate (CaCO_3) and calcium sulphate (CaSO_4). The formation of calcium alumino silicate hydrate and calcium carbonate are from the hydration process while the calcium sulphate detected is the un-reacted raw material. The significant increase in the sorbent BET surface area is believed to be due to the presence of the calcium alumino silicate hydrate compound in the sorbent. The absence of SiO_2 (the main component in coal fly ash) in the diffraction spectrum showed that it reacts completely with CaO to form calcium alumino silicate hydrate. The XRD spectrum, showing the various phases present in the sorbent was comparable with the XRD spectrum reported by Tsuchiai *et al.*, 1995 [1] and Fernandez *et al.*, 1997 [8] on sorbents prepared from $\text{Ca}(\text{OH})_2$, coal fly ash and spent sorbent.

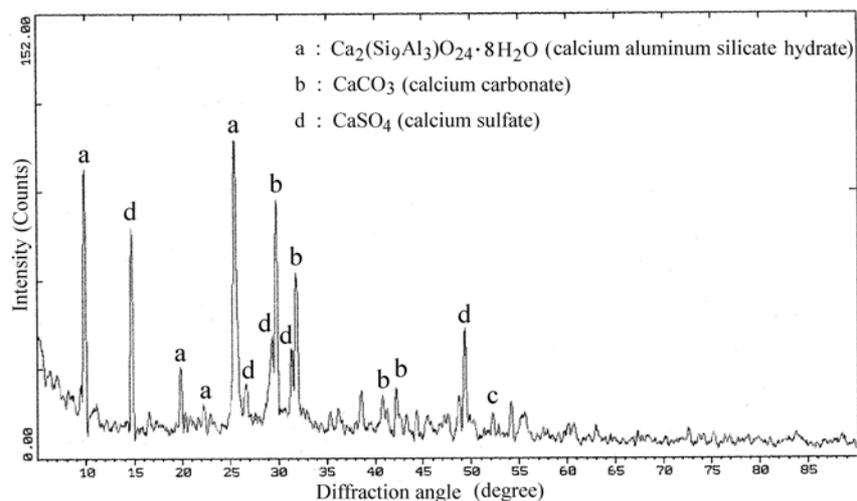


Figure 1. XRD spectrum for sorbent prepared under treatment combination abc.

Hydration experiments based on statistical design were employed according to 2^4 full factorial design to produce sixteen different types of sorbent using sixteen different treatment combinations. These combinations allow the study of the individual influences and their respective interactions of hydration period, amount of fly ash, amount of CaSO_4 and drying temperature towards the sorbents surface area obtained after hydration. The complete design matrix and results are shown in Table 3. Five experiments (S1 to S5) at the centre point of the experimental design were performed in order to determine the experimental error. As the result of the surface area of these 5 runs are quite consistent, single replicate experimental run is essential in this study.

From the data presented in Table 3, it was found that the surface area of the sorbent obtained ranges from 12.9 to 92.7 m²/g, while the highest value of surface area obtained at 92.7 m²/g corresponded to treatment combination abc performed at the following hydration variables; hydration period of 16 hrs., amount of fly ash of 10 g, amount of CaSO₄ of 6 g and drying temperature of 200°C. The effects of individual variables and interaction between variables on the BET surface area of the sorbent were calculated based on 'YATES' algorithm [16] using the response (BET surface area) of the various treatment combinations. The summarized procedure used to calculate effects of the various variables using 'YATES' algorithm is shown in Table 5. The treatment combinations entered into Table 5 were always recorded in standard order [16]. The first half of "Column (1)" is obtained by adding the value of surface area in adjacent pairs under the column "Surface area". The second half of "Column (1)" is obtained by changing the sign of the first entry in each of the pairs in the "Surface area" column and adding the adjacent pairs. For example, in "Column (1)" we obtain $71.6 = 28.4 + 43.2$, $14.8 = -(28.4) + 43.2$ and so on. The values in "Column (2)" are obtained from "Column (1)" just as "Column (1)" is obtained from the "Surface area" column. The values in "Column (3)" are obtained from "Column (2)" and the values in "Column (4)" are obtained from "Column (3)" similarly. The numerical value for the effect of the various variables is then obtained by dividing the data in "Column (4)" by 2^{k-1} where the numerical number 2 represents the level of each variable studied and k is the number of variables studied

(in this case $k = 4$, so $2^{k-1} = 8$). Note that the effect of a particular variable can be obtained from the value in the last column corresponding to the row in the various treatment combinations. For instance, the effect of variable A is 33.2 and the effect of interaction between variable A and B is 16.3.

Table 5. ‘YATES’ algorithm for calculating the effects of the various variables studied.

Treatment combinations	Surface area, m^2/g	Column (1)	Column (2)	Column (3)	Column (4)	Effect of various variables
(l)*	28.4	71.6	156.3	379.9	714.2	-
a	43.2	84.7	223.6	334.3	265.6	33.2
b	26.4	91.4	142.3	123.9	118.0	14.8
ab	58.3	132.2	192.0	141.7	130.6	16.3
c	33.7	64.1	46.7	53.9	117.0	14.6
ac	57.7	78.2	77.2	64.1	36.4	4.6
bc	39.5	71.0	67.9	46.3	63.6	8.0
abc	92.7	121.0	73.8	84.3	22.6	2.8
d	24.3	14.8	13.1	67.3	-45.6	-5.7
ad	39.8	31.9	40.8	49.7	17.8	2.2
bd	12.9	24.0	14.1	30.5	10.2	1.3
abd	65.3	53.2	50.0	5.9	38.0	4.8
cd	28.9	15.5	17.1	27.7	-17.6	-2.2
acd	42.1	52.4	29.2	35.9	-24.6	-3.1
bcd	30.2	13.2	36.9	12.1	8.2	1.0
abcd	90.8	60.6	47.4	10.5	-1.6	-0.2

**(l)- All the variables are at low level*

Based on the calculated data in Table 5, the effect of the various variables can now be plotted in a probability plot. Table 6 shows the data required for the probability plot while the probability plot itself is given in Fig. 2. From Fig. 2, it is seen that a straight line can be drawn passing through most of the points on the plot except for points corresponding to variables

A, B, C and AB. This result indicates that variables A, B, C and the interaction between variable A and B are the significant variables that influence the BET surface area of the sorbent. It also indicates that there is no high order interaction between the experimental variables that exerted significant influence on the BET surface area of the sorbent. A linear model was developed

Table 6. Tabulated data for the probability plot.

Order (j)	Variable	Effect of various variable	$P_j = (j-0.5)/15$
15	A	33.2	0.9667
14	AB	16.3	0.9000
13	B	14.8	0.8333
12	C	14.6	0.7667
11	BC	8.0	0.7000
10	ABD	4.8	0.6333
9	AC	4.6	0.5667
8	ABC	2.8	0.5000
7	AD	2.2	0.4333
6	BD	1.3	0.3667
5	BCD	1.0	0.3000
4	ABCD	-0.2	0.2333
3	CD	-2.2	0.1667
2	ACD	-3.1	0.1000
1	D	-5.7	0.0333

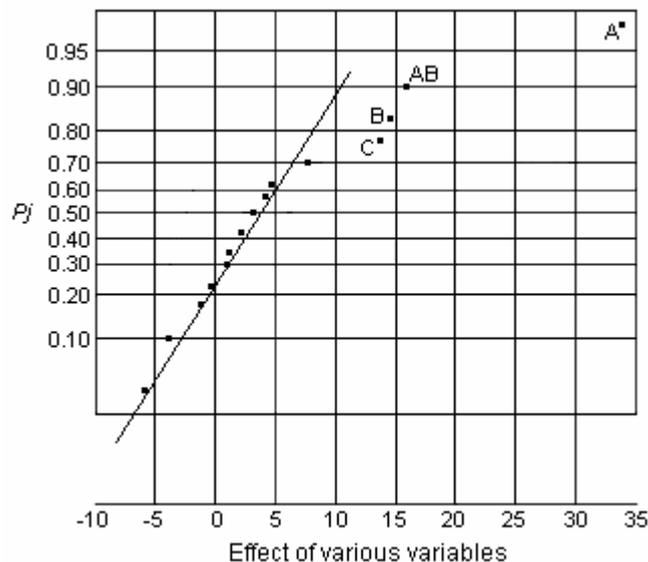


Figure 2. Probability plot for the effect of various variables.

based on these four significant variables. The coefficient for the variables is taken from the numerical values of the effect of the various variables tabulated in Table 5. The linear model for the BET surface area is given in Eq. (2):

$$\text{Surface area (m}^2\text{/g)} = 44.6 + \left(\frac{33.2}{2}\right)A + \left(\frac{14.8}{2}\right)B + \left(\frac{14.6}{2}\right)C + \left(\frac{16.3}{2}\right)AB \quad (2)$$

where the first term in Eq. (2) corresponds to the average BET surface area for all the sorbents prepared under various treatment combinations as tabulated in Table 3 while A, B, C and AB are the significant variables which take the value of 1 or -1 based on the treatment combinations as also tabulated in Table 3. For instance, in order to calculate the predicted BET

surface area of sorbent obtained under treatment combination a, the terms A,B and C in Eq. (2) are replaced by 1, -1 and -1 respectively. Note that in Eq. (2), the coefficient of each variable is obtained by dividing the effect of the respective variable by two as in the calculation procedure, the effect of a variable represents a change from -1 to 1, or a change of two units (from -1 to 0 and 0 to 1). A positive sign in front of the terms indicates the synergistic effect of that particular variable on the surface area of the sorbent. Fig. 3 shows the experimental BET surface area versus predicted BET surface area using the model equation developed. A line of unit slope or a line of perfect fit with points corresponding to zero error between experimental and predicted values is also shown in Fig. 3. These plots therefore visualize the performance of the model in an obvious way. From Fig. 3, it can be seen that the model developed predicts the BET surface area of the sorbent quite accurately.

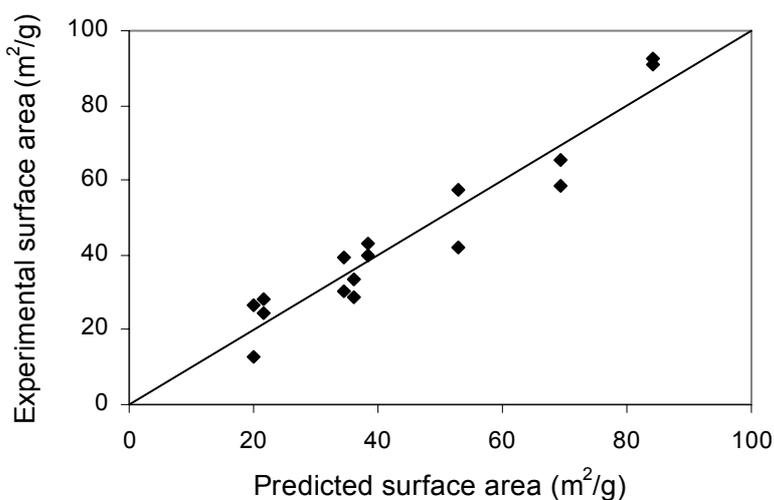


Figure 3. Experimental versus predicted BET surface area.

The coefficients of determination (R^2) for Eq. (2) was determined as 0.914 and the absolute average percentage deviation (AAPD) between calculated and experimental BET surface area were found to be less than 10%. From these statistical tests (the high value of coefficients of determination and the low value of absolute average percent deviation) together with the plot presented in Fig. 3, it was demonstrated that the mathematical model equation provided quite an accurate description of the experimental data, indicating that it was successful in capturing the correlation between the significant sorbent preparation variables to the surface area of the resulting sorbent.

For further diagnostic checking of the model, analysis of variance (ANOVA) is used. Table 7 presents the results of the analysis of variance calculated using Design-Expert 6.0.6 software. Based on 1% level of significance, the mathematical model as in Eq. (2) was tested to be significant as the computed F value (29.3) is much higher than the theoretical $F_{0.01(4,11)}$ value (5.67), indicating that the model is reliable in predicting the sorbent surface area. Apart from that, it is also shown that the four variables (A,B,C and AB) are tested (based on 1% level of significance) to be the only significant terms in the model as the individual computed F value is higher than the theoretical F value. Thus, it can be concluded that at a confidence level of 99%, variable A, B, C and as well as the interaction between variable AB are the only significant variables that effect the BET surface area of the sorbent.

Table 7 : Analysis of variance (ANOVA) for the sorbent BET surface area.

Source of variance	Sum of squares	Degrees of freedom	Mean square	F
A	4409.0	1	4409.0	71.8
B	870.3	1	870.3	14.2
C	855.6	1	855.6	13.9
AB	1066.0	1	1066.0	17.4
Model	7200.9	4	1800.2	29.3
Residual	675.6	11	61.4	-
Total	7876.5	15	-	-

Eq. (2) clearly shows that the coefficient for hydration period (variable A) is the highest among the other three variables. Thus, it is possible to conclude that the effect of this variable on the surface area of sorbent is the strongest. It was reported that the dissolution rate of SiO_2 present in the ash is the rate limiting step for the formation of calcium aluminum silicate hydrate compounds in the sorbent [14]. This is in agreement with our results because as the hydration period proceeds longer, more SiO_2 elutes out from the ash to react with CaO to form calcium aluminum silicate hydrate compounds and thus gives a positive effect on the sorbent surface area.

The amount of fly ash and CaSO_4 (variable B and C) used in the preparation mixture has also a positive effect on the surface area of the sorbent. In fact, the effect of these two variables are similar as the magnitude of the coefficients are quite close. The amount of fly ash generally determines the

amount of SiO_2 available in the preparation mixture to react with CaO to form the active species during the pozzolanic reactions. Thus a higher amount of fly ash is an indication that more SiO_2 is present in the preparation mixture for it to react with CaO to form the active species and thus give a positive effect on the surface area of the sorbent. The positive effect of CaSO_4 on the surface area of sorbent prepared from coal fly ash was also reported by Ishizuka *et al.*, 2001 [7] However, in his work, Ca(OH)_2 is used as the raw material instead of CaO . It was reported that this phenomena is due to the role played by CaSO_4 where it promotes the formation of calcium silicate compounds by suppressing crystal growth of Ca(OH)_2 to keep the reactivity of the Ca(OH)_2 towards coal fly ash. Thus it can be concluded that CaSO_4 has the same effect on CaO as it has on Ca(OH)_2 . Apart from this, the interaction effect between variable A and B also exerted significant positive effect on the surface area of the sorbent. In other words, if one of the variables is changed with respect to another one, it will have a considerable effect on the total surface area of the sorbent.

Conclusion

Sorbents for dry flue gas desulphurization were prepared from coal fly ash, CaO and CaSO_4 using water hydration. From the experiment, it was concluded that there is a strong influence of the following sorbent preparation variables; hydration period, amount of fly ash and amount of CaSO_4 , towards the sorbent BET surface area while its dependence on the drying

temperature is negligible. The linear model developed for predicting the surface area of the sorbent was found to provide an accurate description of the experimental data, indicating that it was successful in capturing the correlation between the significant sorbent preparation variables to the surface area of the resulting sorbent.

Acknowledgement

The authors would like to thank the ASEAN University Network/Southeast Asia Engineering Education Development Network (AUN/SEED-Net), JSPS-VCC (Program on Environmental Science, Engineering and Ethics), Ministry of Science, Technology and Environment Malaysia (Project No. 08-02-05-2040EA001) and Universiti Sains Malaysia (USM short term grant) for the funding and support on this project.

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