

REMOVAL OF SULFUR DIOXIDE FROM FLUE GAS USING ABSORBENT PREPARED BY WATER AND STEAM HYDRATION

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ABSTRACT

Active absorbent for flue gas desulfurization was prepared from coal fly ash, calcium oxide (CaO) and calcium sulfate (CaSO₄) by hydro-thermal process; steam and water hydration. The absorbents were examined and compared for its micro-structural properties. The experiments were conducted based on Design of Experiments (DOE) according to 2⁴ factorial design. The effect of various absorbent preparation variables such as hydration period (Factor A), ratio of CaO to fly ash (Factor B), amount of calcium sulfate used (Factor C) and drying temperature (Factor D) towards the BET surface area of the absorbent were studied. The BET surface area of the absorbent was in the range of 12.9-169.3 m²/g. Fisher's test showed that there is a strong influence of factor A, B and D towards the absorbent surface area, while its dependence on factor C is negligible. Comparison between absorbents prepared from water and steam hydration showed that the BET surface area of absorbents prepared from water hydration gives a higher surface area, but at a lower rate. The optimum BET surface area for the prepared absorbent 169.3 m²/g, was obtained at the following absorbent preparation variables using water hydration; hydration period of 24 hours, ratio of CaO to fly ash of 1:2, CaSO₄ amount of 3g and drying temperature of 200°C.

Key words : *absorbent, BET surface area, coal fly ash, Design of Experiments, desulfurization.*

1. INTRODUCTION

In an effort to protect the environment, industrial operations are required to reduce its emission

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of air pollutants. Among air pollutants recognized world wide is sulfur dioxide (SO₂). SO₂, with an acidic and toxic characteristics, is the primary cause of acid rain and can cause harmful effect towards vegetation and human health. The primary source of SO₂ comes from boilers running on solid and liquid fuels such as those used by power plants, petrochemical, oleochemical, plastics, fabrics and many more other industries. This is due to the sulfur compound present in the fuel, which will react with air to form SO₂ during the combustion process in the boiler. The SO₂ formed is then released to the environment through the boiler's flue gas. The concentration of SO₂ released could easily reach 2500 ppm¹, whereas the permissible concentration allowed by most environment control regulation around the world is only about 500 ppm². Therefore, in order to reduce the emission of SO₂, flue gas from the boiler has to be treated with appropriate yet affordable desulfurization technology before being emitted from the chimney to the environment.

Presently, different types of flue gas desulfurization (FGD) units are being operated. A wet-type FGD unit based on a limestone-gypsum method is most widely used and suitable for large-scale boilers such as those installed in coal or oil-fired power stations³. The wet-type FGD process has the advantage of using inexpensive limestone as a neutralizing agent, to produce commercial gypsum and to remove sulfur dioxides at high efficiency. On the other hand, disadvantages of this process include the large space needed for installation, the large volume of water required and the high capital and operating expenses. In view of these problems, researchers are currently shifting their focus in developing dry desulfurization process to be applicable to the power stations in the region where water supply is not sufficient and to reduce the cost in installing and operating desulfurization unit in small and medium scale industries.

Utilization of coal fly ash in the preparation of absorbent for dry FDG system presents advantages both economically and environmentally. Coal fly ash is a waste product from coal power plants that has not much commercial value. At the moment, most of the ash produced has to be disposed off, either in landfills or ash ponds. This method of ash disposal requires a lot of land area, which is not easily available in urban areas. On the other hand, when coal fly ash is used to make the absorbent, it will be converted into gypsum upon absorbing SO₂. In contrast to coal fly ash, gypsum (CaSO₄.2H₂O) is a commercial product used in the building industries. It can also be used to manufacture deodorant, coagulating agent and fertilizers⁴. Apart from that, utilization of the waste product as the absorbent to control the pollution cause by the same process seems to be very economical.

The active absorbent is due to the pozzolanic reaction that takes place between fly ash and calcium oxide/calcium hydroxide which yield highly hydrated compounds such as calcium silicates and calcium aluminates which has been thought to be responsible for the improvement in solid utilization (compared to using calcium oxide/calcium hydroxide as the absorbent individually)⁵. The influence of the absorbent micro-structural properties such as the BET specific surface area, which is one of the major parameters affecting the efficiency of the absorbent in removing SO₂, has been mentioned by a number of researchers^{1,3,5-10}. Recently, Ishizuka *et al.* 2001¹ reported that the changes in the activity of the absorbent are in accordance with the increase of BET specific surface area of the absorbent. High surface area in the

absorbent make calcium ions more accessible to SO_2 molecules. Apart from that, it also provides the absorbent with the ability to carry more water molecules on the absorbents outer surface which will then promotes the reaction between SO_2 and calcium to occur.

Generally, hydro-thermal hydration process is being used to prepare highly active absorbents from coal fly ash. For pressure hydration, absorbents with a BET specific surface area of 2.5-64.3 m^2/g were obtained⁵. On the other hand, for steam hydration and water hydration, values of 24 m^2/g ⁵ and 18-73 m^2/g were obtained respectively^{3,7}. It can be seen that the products of the pozzolanic reactions differs in each case depending on the methods of absorbent preparation as well as the fly ash composition and absorbents preparation parameters. However, it is clear that the products of the pozzolanic reaction are highly porous material with higher surface area compared to the reactant. Many researchers have reported the preparation of absorbents from various techniques, but reports on comparison between techniques to prepare the absorbents are still limited. Ishizuka et. al. 2001¹ reported that, for steam hydration preparation method, absorbents prepared from calcium oxide instead of calcium hydroxide exhibits higher activity in SO_2 removal.

In the present study, absorbents prepared from coal fly ash using steam hydration and water hydration were investigated. The effect of various absorbent preparation variables such as hydration period (factor A), ratio of CaO to fly ash (factor B), amount of calcium sulfate used (factor C) and drying temperature (factor D) towards the BET specific surface area of the absorbent were studied. Design of Experiments based on full factorial design of 2^4 was used in the present study to establish the significance of each independent variables and interaction between variables using Fisher's Test¹³. Upon identification of the significant variables, the range of significant variables studied will be extended for optimization purposes. Comparison between the BET surface area of absorbent prepared from both preparation methods was also carried out in this work.

2. EXPERIMENTAL

The absorbents were prepared from calcium sulfate (CaSO_4), calcium oxide (CaO) and coal fly ash. The calcium sulfate used was of reagent grade, calcium sulfate hemihydrate (BDH Laboratory Supplies, England), whereas the calcium oxide used was of laboratory grade (BDH Laboratory Supplies, England). The coal fly ash was supplied by Kapar Power Plant, Malaysia of Tenaga Nasional Berhad. The coal fly ash has the following composition: SiO_2 42.0%, Al_2O_3 19.0%, CaO 4.6%, Fe_2O_3 9.6%, MgO 2.1%, NaO 1.6% and ignition loss 21.1%.

Standard procedure to prepare the absorbent is as follows. For steam hydration³, to prepare 20 g of the absorbent (dry weight), a mixture of coal fly ash (50% of the total used) and calcium oxide was mix with 28-35 % of water at 70°C for 5 minutes. Then calcium sulfate and the rest of the coal fly ash were added into the slurry and kneading was continued for 10 minutes. The kneaded materials were then extruded to form pellets and were subjected to hydrothermal treatment with steam for a specific period of time followed by drying for 2 hours at a specific temperature. The dried pellets were then crushed and sieved through a $60\ \mu\text{m}$ mesh. For water

hydration⁷, to prepare 20g of absorbent, calcium oxide was added into 100 ml of water at 65°C. Upon stirring, the temperature of the slurry will eventually increase to about 80°C. Fly ash and calcium sulfate were then added into the slurry simultaneously. The slurry was then heated at about 95°C for a specific period of time for the hydration process to occur. After the hydration process, the resulting slurry was filtered and dried at a specific temperature for 2 hours. The

Table 1 : *Experimental design matrix and results*

Solid code*	Experimental variables				BET surface area (m ² /g)	
	Hydration period, t (hrs)	Ratio (weight) of CaO to fly ash, C:F	Amount of CaSO ₄ used, m (g)	Drying temperature, T(°C)	Steam hydration	Water hydration
S1/S18	4	2:1	3	200	30.1	33.7
S2/S19	10	2:1	3	200	46.5	57.5
S3/S20	4	1:2	3	200	58.9	39.5
S4/S21	10	1:2	3	200	86.4	92.7
S5/S22	4	2:1	6	200	21.6	28.9
S6/S23	10	2:1	6	200	24.0	42.1
S7/S24	4	1:2	6	200	54.1	30.2
S8/S25	10	1:2	6	200	59.7	90.8
S9/S26	4	2:1	3	500	18.9	28.4
S10/S27	10	2:1	3	500	42.0	43.2
S11/S28	4	1:2	3	500	42.0	26.4
S12/S29	10	1:2	3	500	54.8	58.3
S13/S30	4	2:1	6	500	19.7	24.3
S14/S31	10	2:1	6	500	21.2	39.8
S15/S32	4	1:2	6	500	33.4	12.9
S16/S33	10	1:2	6	500	58.4	65.3
S17-I/ S34-I	7	1:1	4.5	350	45.5	59.9
S17-II/ S34-II	7	1:1	4.5	350	57.0	54.3
S17-III/ S34-III	7	1:1	4.5	350	51.7	56.2
S17-IV/ S34-IV	7	1:1	4.5	350	56.2	61.0
S17-V/ S34-V	7	1:1	4.5	350	53.6	58.2

* S1 to S17 are for absorbents prepared from steam hydration while S18 to S34 are for absorbents prepared from water hydration.

amount of chemicals used and experimental parameters are given in Table 1.

The absorbents were analyzed for its BET specific surface area using Autosorb 1C Quanta-chrome¹¹. The value of BET specific surface area were calculated using a software (Micropore 2.46) which was interface on-line with the Autosorb instrument. Prior to analyzing the sample, it was degassed at 200°C for at least 8 hrs. XRD patterns were recorded on a Philips PW 1820 system for the powdered samples with Cu-K α radiation in the range of diffraction angle (2θ) 1.5°-90° at a sweep rate of 1 deg/min¹².

3. RESULTS AND DISCUSSION

BET surface area for coal fly ash, calcium oxide (CaO), S21 (maximum value of absorbent BET surface area prepared from water hydration)⁹ and commercial absorbents (typically calcium hydroxide)⁹ are given in Table 2. The significant increase in the specific surface area of the hydrated coal fly ash absorbent 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 hydrated absorbent prepared has a mixture of cylindrical granular grains, which consists of predominantly calcium silicate, calcium alumino sulfate and CaO. This calcium compounds are believed to be the main component in absorbing SO₂. The significant increase in the surface area of the hydrated absorbent also indicates the formation of absorbents with a higher structural porosity as compared to the regular CaO. The high surface area of a solid absorbent is an important parameter in the solid-gas adsorption process as it may

Table 2 : BET surface area of raw materials and hydrated absorbents

Material	BET surface area (m ² /g)
Uncalcined coal fly ash	1.46
Calcium Oxide (CaO)	1.52
Prepared absorbent (S21)	92.7
Commercial absorbent	16.2

be one of the principal factors that determine the overall reactivity.

The formation of calcium silicate and calcium alumino sulfate (Ca₄Al₂SO₁₀.12H₂O) was confirmed by the X-Ray Diffraction (XRD) patterns obtained. Figure 1 (a) and (b) shows the XRD pattern for absorbents prepared from steam hydration (S4) and water hydration (S21), respectively. The peaks characteristic for calcium silicate was observed at $2\theta = 29.22^\circ$ in the XRD pattern for both samples. However, the peak for calcium alumino sulfate at $2\theta = 9.93^\circ$ and 19.9° was only observed in the XRD pattern for the absorbent prepared from water hydration (S21). This shows that calcium alumino sulfate was not formed in the absorbent prepared from steam hydration method due to the limitation of water availability during the preparation method. The limitation of water cause the raw materials not being able to react extensively to form calcium

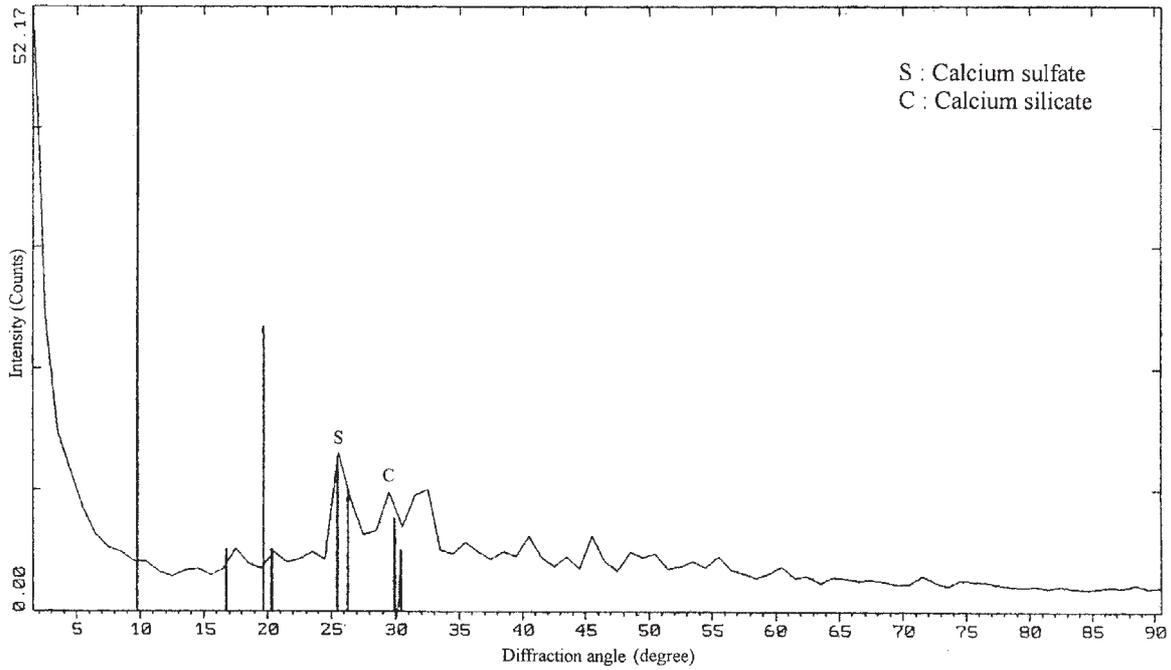
alumino sulfate which is a more complex structure compared to calcium silicate. On the other hand, the peak for calcium sulfate was detected in both of the absorbents at $2\theta = 25.4\theta$. The detected calcium sulfate is the un-reacted calcium sulfate. The XRD patterns, showing the presence of calcium silicate, calcium alumino sulfate and calcium sulfate in the absorbents were comparable with the XRD patterns reported by Tsuchiai *et. al.* (1995)⁶ and Ishizuka *et. al.* (2001)¹ on absorbents prepared from Ca(OH)₂, coal fly ash and used absorbent.

Hydration experiments based on DOE were employed according to 24 factorial design to produce 16 different types of absorbents for each absorbent preparation method as shown in Table 1. Absorbents prepared using steam hydration were coded as S1 to S17 while absorbents prepared from water hydration were coded as S18 to S34.

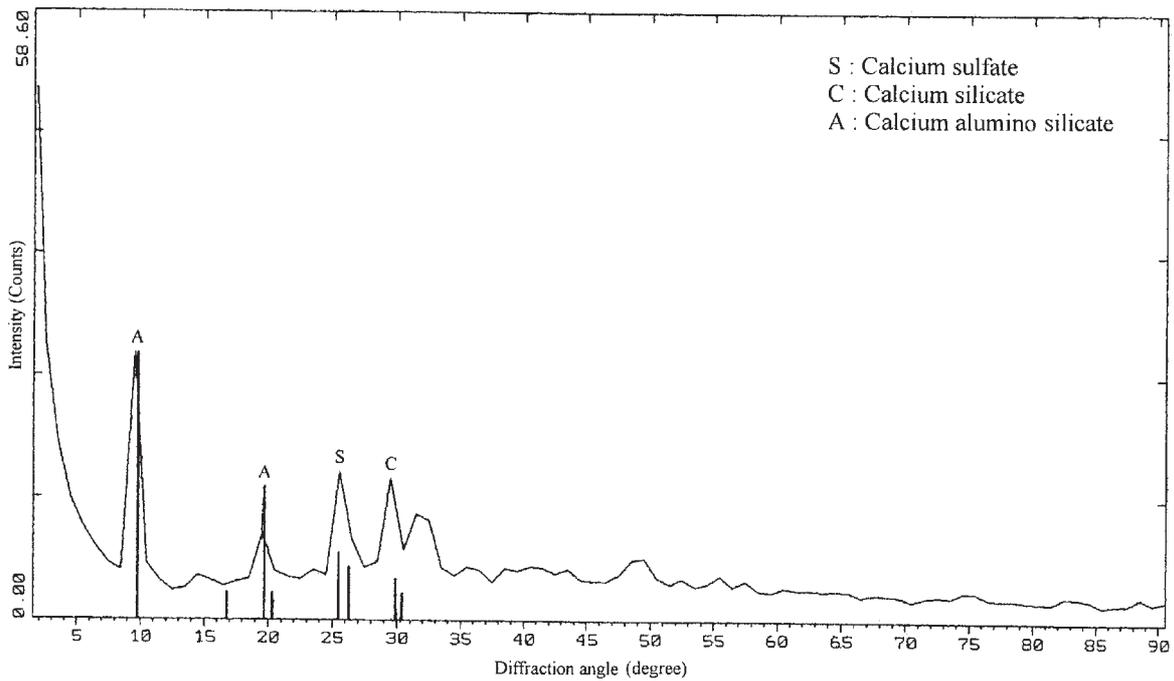
All the 42 absorbents obtained were tested for its BET surface area. These combinations allow the studying of the influences of individual experimental variables and their respective interactions towards the absorbent BET surface area. The four absorbent preparation variables studied in this work were coded as factor A for hydration period, factor B for ratio of calcium oxide (CaO) to fly ash, factor C for amount of calcium sulfate (CaSO₄) added to the absorbent and factor D as the drying temperature. Interaction between factors such as between hydration period and ratio of CaO to fly ash was coded as AB. The range of the experimental variables studied are as presented in Table 1. Experiment 17-I to 17-V and 34-I to 34-V at the center point of the experimental design were performed in order to determine the experimental error. From the results, it can be seen that the BET surface area obtained were quite consistence indicating that a single replicate experiment is essential in this study.

In relation to the results of BET surface area tabulated in Table 1, it was found that the value obtained ranged from 18.9-86.4 m²/g and 12.9-92.7 m²/g for absorbents prepared from steam and water hydration, respectively. It was also observed that the optimum BET surface area obtained for absorbent prepared from both preparation method was obtained at the following absorbent preparation parameters ; at a hydration period of 10 hrs., CaO to fly ash ratio of 1:2, 3g of CaSO₄ used and drying temperature of 200°C. Apart from that, from the results presented in Table 1, it was observed that higher hydration period and higher fly ash ratio resulted in absorbents with higher surface area for both preparation methods. Longer hydration period will ensure enough time for reactions between reactants to be completed while higher fly ash ratio will ensure that the calcium compound in the absorbent is fully reacted to form the highly porous material (calcium silicate and calcium alumino sulfate) in the absorbent.

On the other hand, increasing the drying temperature has a negative effect on the absorbents surface area. It was reported previously that by increasing the drying temperature of the absorbent, the content of calcium carbonate (CaCO₃), which has a lower surface area, will increase⁶. One possible explanation for the decrease in surface area is due to the decomposition of porous material back to CaO, which will then react with carbon dioxide (CO₂) to form CaCO₃. Increasing the amount of CaSO₄ used in preparing the absorbent also has a negative effect on the absorbents surface area. For absorbents prepared from steam hydration, this trend is similar to those reported by Ishizuka *et. al.* (2001)¹. When calcium sulfate is added with CaO during the



(a)



(b)

Figure 1 : XRD pattern for absorbent prepared from (a) steam hydration and (b) water hydration

slaking step, it will cover the outer surface of calcium hydroxide and thus reduces its reactivity towards coal fly ash to form calcium silicate¹. However, the trend obtained for absorbents prepared by water hydration is not in agreement with those reported by Ishizuka et. al. (2001)¹. It was reported that the surface area of absorbent increases with the addition of CaSO₄.

This is due to the role played by CaSO₄ where it promotes the formation of calcium silicate by suppressing crystal growth of calcium hydroxide to keep the reactivity of the calcium hydroxide towards coal fly ash. In this work, it was however found that the addition of CaSO₄ reduces the surface area of the absorbents prepared. This indicates that, as in the case for absorbents prepared by steam hydration, addition of CaSO₄ during preparation of absorbents using water hydration will also cover the outer surface of calcium hydroxide and thus reduces its reactivity towards coal fly ash to form calcium silicate.

In order to examine the statistical significance of each independent variables and interaction between variables on the BET surface area of the prepared absorbent, Fisher's test is applied on

Table 3 : Fisher's Test on the various effects of absorbent preparation variables on the BET surface area

(a) steam hydration

Source of variance	Sum of squares	Degree of freedom	Mean square	F _o
Model	5224.18	6	870.70	21.5a
A	814.3	1	814.3	20.1a
B	3126.5	1	3126.5	77.1a
C	479.6	1	479.6	11.8a
D	519.8	1	519.8	12.8a
AC	127.1	1	127.1	3.1b
BD	156.9	1	156.9	3.9b
Residual	364.9	9	40.5	-
Total	5589.1	15	-	-

(b) water hydration

Source of variance	Sum of squares	Degree of freedom	Mean square	F _o
Model	7193.1	4	1798.3	29.4a
A	4400.3	1	4400.3	71.9a
B	873.2	1	873.2	14.3a
D	850.6	1	850.6	13.9a
AB	1069.0	1	1069.0	17.5a
Residual	672.9	11	61.2	-
Total	7866.0	15	-	-

^abased on 99% confidence level

^bbased on 90% confidence level

the results of the Design of Experiment¹³. Tables 3 (a) and (b) shows the result of the Fisher's test calculated using Design-Expert Software 6.0¹³ for absorbent prepared from steam and water hydration, respectively. The significant variables are identified based on 99% and 90% confidence level. Only the results of the significant variables are shown. From Table 3 (a), it can be concluded that Factor A, B, C and D as well as the interaction between factors AC and BD are the significant factors that effect the BET surface area of the absorbent prepared using steam hydration. The significance of Factor A, B, C and D in affecting the absorbent surface area are based on 99% confidence level while for Factor AC and BD, they are based on 90% confidence level. On the other hand, based on Table 3 (b), for absorbents prepared from water hydration, the BET surface area of the prepared absorbent are significantly influenced by Factor A, B and D as well as the interaction between factors AB. The significant of all these factors are based on 99% confidence level. It can also be concluded that there are no high order interaction between factors that affect the absorbent surface area for absorbents prepared by both preparation methods. Apart from that, from these results, it can also be seen that for absorbents prepared from both experimental method, Factor A or hydration period and Factor B or ratio CaO to fly ash are the most influential factor among the significant factors, as they have the highest F value.

Based on the results presented earlier, it was found that higher hydration period and higher fly ash ratio resulted in absorbents with higher surface area for both preparation methods. However, the optimum surface area obtained from Design of Experiment was found to lie at the extreme point (maximum hydration period of 10 hrs and ratio of CaO to fly ash of 1:2). Hence in order to complement the results and to obtain the optimum surface area of the absorbents more accurately, the range of Factor A and B were further studied by extending its range.

Figure 2 shows the effect of hydration period on the surface area of absorbents prepared from steam and water hydration (the absorbents were prepared using the following preparation vari-

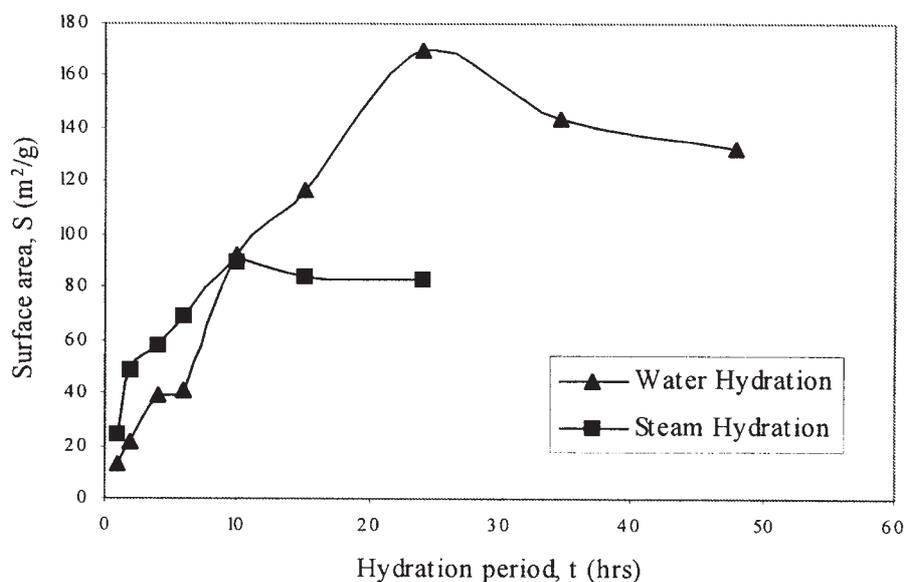


Figure 2 : Effect of hydration period on absorbents BET surface area

ables ; ratio of CaO to fly ash of 1:2, CaSO₄ amount of 3g and drying temperature of 200°C). The range of hydration period studied was extended from 4-10 hours to 4-50 hours. In steam hydration, the optimum surface area of the absorbent was attained at a hydration period of about 10 hours. The surface area slightly decreases at a longer hydration period. Compared to steam hydration, the surface area of absorbent prepared from water hydration achieved its maximum value at a hydration period of 25 hours. The optimum surface area for absorbent prepared from water hydration (170 m²/g) is almost double of that absorbent prepared from steam hydration (85 m²/g). This could be attributed to the following reason. In water hydration, all the raw materials are well mix through-out the whole hydration period, thus giving a complete reaction to produce highly porous material, but in steam hydration, the absorbent was first formed into pellets prior to hydration. Upon exposure to steam, only the outer layer of the pelleted absorbent reacted to form the highly porous material. The inner part of the pellet remain un-reacted, thus limiting the amount of highly porous material formed. However, this could be overcome by using a smaller pellet or by using pressurized hydration. Figure 2 also shows that the surface area of absorbent prepared from steam hydration exceeds the surface area of absorbent prepared from water hydration for hydration period less than 10 hours. This shows that the rate of reaction of the formation of calcium silicate and calcium alumino sulfate (porous material that gives high surface area in the absorbent) in the absorbent prepared by steam hydration is faster than water hydration for shorter hydration period.

Figure 3 shows the effect of CaO to fly ash ratio on the absorbents surface area (the absorbents were prepared using the following preparation variables ; hydration period of 4 hrs, CaSO₄ amount of 3g and drying temperature of 200 (C). From Figure 3, it is observed that the optimum surface area for absorbents prepared by both preparation methods is obtained at a CaO to fly ash ratio of about 1 : 1.9. At lower fly ash ratio, there might be not enough silica and alumina

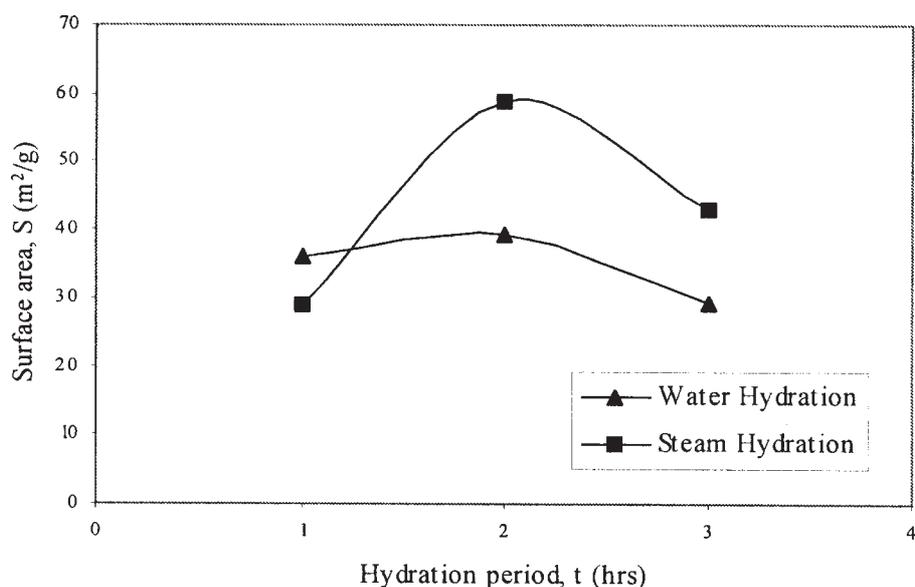


Figure 3 : Effect of CaO to fly ash ratio on absorbents BET surface area

to react with calcium (Ca) species to form porous material in the absorbent while at a higher ratio, there might be too much inert present in the absorbent until it prevents the hydration reaction from occurring completely.

4.0 CONCLUSIONS

Absorbents for dry flue gas desulfurization were prepared from calcium oxide, coal fly ash and calcium sulfate using steam and water hydration. The effect of various absorbent preparation variables such as hydration period, ratio of CaO to fly ash, amount of calcium sulfate used and drying temperature towards the BET specific surface area of the absorbent were studied using Design of Experiments (DOE). From the experiment, it was concluded that there is a strong influence of the following absorbent preparation variables ; hydration period, ratio of CaO to fly ash and drying temperature, towards the absorbent BET surface area while its dependence on the amount of CaSO₄ used is negligible. Apart from that, it can also be concluded that hydration period and ratio of CaO to fly ash are the crucial factors that must be controlled to obtain highly porous absorbent (absorbents with high surface area). Comparing the results of both preparation methods, it was found that absorbents prepared from water hydration gives a higher surface area compared to steam hydration. However, the rate of formation of porous material that gives higher surface area in the absorbent is slower in water hydration compared to steam hydration.

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NOMENCLATURE

a	based on 99% confidence level
A	Hydration period
AB	Interaction between hydration period and ratio of CaO to fly ash
AC	Interaction between hydration period and amount of CaSO ₄ used
b	based on 90% confidence level
B	Ratio of CaO to fly ash
BET	Brunauer-Emmett-Teller
BD	Interaction between ratio of CaO to fly ash and drying temperature
C	Amount of CaSO ₄ used
D	Drying temperature
DOE	Design of experiment
FGD	Flue gas desulfurization