REMOVING BASE MIST FROM FLUE GAS EMISSION

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Abstract— **Air pollution is a mixture between substances from natural gas with man-made where almost all factories contribute to harmful gases as their waste in the production line. This problem does not only effect human but also to animal and plants. Most studies on treating air pollution are only focus on one compound in the pollution and usually on acid gases compound. However, there are some base compound exists in the atmosphere creating air pollution. In order to overcome this problem, this study has been conducted which aiming to study the removal of various base gas using sulfuric acid solution and to investigate the effect of concentration of sulfuric acid and contact time toward base gas. This research will focused in three types of base, which are ammonia, triethylamine, and diethyl ether. The base gases were passed through various concentration of sulfuric acid with different time contact. Ammonia was removed with 0.05M, 0.1M and 0.5M of sulfuric acid and time taken of 5, 10, 15 and 20 minutes. Triethylamine was removed with 0.01M, 0.05M, 0.1M and 0.5M of sulfuric acid together with time of 10, 15, 20 minutes. Lastly, diethyl ether was removed with time contact of 5, 10, 15 and 20 minutes with 0.01M, 0.05M, 0.1M and 0.5M of sulfuric acid. The data were analyse using Gas Chromatography. Based on the analysis, higher concentration of sulfuric acid with longer contact time shows the most removal of base emission for all type of base. The best concentration and time for removal were the lowest and the shortest. For ammonia, 0.5M of sulfuric acid at 5 minutes achieved 100% efficiency removal. 100% efficiency of removal triethylamine achieved 0.1M for 5 minutes. Lastly, the best condition for diethyl ether is 0.1M of sulfuric for 5 minutes of contact time.**

Keywords— Sulfuric Acid, Scrubber, Base Gas, Removal Efficiency

1. INTRODUCTION

Air pollution problem is not happening in certain country but all over the world. This is because, as the rising in technology, more production demand increase therefore create more harmful gases to the environment. There are various type of gaseous air pollution such as sulfur dioxide (SO), nitrogen oxide (NO), nitrogen dioxide (NO_2), carbon monoxide (CO), ammonia, volatile organic compounds (VOCs) and persistent organic pollutants (POPs). This gaseous air pollution are from anthropogenic sources which means from hand man made air pollution. For example, livestock farming contribute the most emission since decomposing of animal waste and urea acid produce ammonia. Volatile organic compound from road transport, solvent use and production process. The gaseous can be divided into acid gas and base gas, which depend on their characteristics.

Air pollution today are at crucial state. A lot of human and animals were effected of by these phenomena. Compared to acid gas, alkaline gas emission are lesser however, it also bring negative impact to human, animal and plant. The dangerous of base gas have been documented in a lot of study and a lot of accident had occur regarding on base gas. Alkaline gas can react with water in the air and comes down to earth as alkaline solution. Drinking alkaline water at pH 8.8 can deactivate pepsin and reduce stomach acid [1]. For healthy people, lower acid stomach can create many problem.

Ammonia is one type of base gas that produce sharp odor and large concentration which can give immediate hazard to life. OSHA exposure limit for ammonia gas is 35 ppm in air for 15 minute in a day [2]. Ammonia could affect aquatic life, in long term of exposure can change the gill structure of a fish and oxygen uptake can seriously impaired [3]. There are many invention of technologies had been develop in order to control ammonia emissions. Wet scrubber is known as the most effective pollution control technology with 99% efficiency [4] [5]. There are several studies used acid scrubbers in ammonia removal, however, these studies takes long times with high air flow of ammonia [6][7][8]. Removal of ammonia using reverse osmosis water are only 85.4% removal with time 210 minutes while tap water only shows 64.6% removal [9]. This shows it is less efficient than acid scrubber is.

Amine is also a type of base, which are derivatives from ammonia. Excess amount of amines in the air creates environmental problem especially amines characteristics produce odor. Amines are carcinogen towards human which can creates human tumor and urinary bladder cancer [10]. It also have low molecular weight, which can cause malfunction of kidney by block of voltage-dependent calcium channel in human embryonic kidney cells and can cause asthma [11]. Study on amine removal are evasive since amine are usually used as absorbent to remove acid gases such as hydrogen sulfide and carbon dioxide. In research conducted by [12] removal of dimethyl amine using sulfuric acid, it can achieve up to 95%. This shows that acid scrubber are able to remove amine.

Other type of base gas are diethyl ether. Diethyl ether also brings damage to living things. Diethyl ether are more soluble in blood than water because of it hydrogen bond with water [13]. In mid 1800s, it has been used as inhalation anesthetic agent. Patient exposed to diethyl ether will increase of alkaline phosphatase and aspartate aminotransferase and reducing of serum albumin in liver make the liver function abnormally [14]. This effect also is a similarities effect on animals experimental varies fatty infiltration to central necrosis and liver damage [13]. Diethyl ether is easily vaporize compound with highly flammable and it is usually treat as hazardous waste disposal. There are study on controlling diethyl ether vapor using activated carbon with 0.004 ppm it can achieve 99.7% removal [15].

Variation of method in removing these gases make it harder to remove simultaneously, thus, this research project are focusing on removal of base gas using only one solution. This will help industries that produce these base gases to remove them at the same time, hence, reduce the cost. There are three types of base used, which are ammonia, amine, and diethyl ether. The removal of gases will be absorb by one type of solution, which is sulfuric acid; hence, absorption (wet scrubbing) method will be use.

Sulfuric acid is one of type of inorganic acid, since it nature is acidic, it can easily neutralize base compound. Sulfuric acid is the common acid used in air scrubber to remove ammonia because it is non-volatility and low cost [5]. It can directly converted ethylene oxide to ethylene glycol and expected to absorb ethylene oxide faster than hydrochloric acid [15]. Sulfuric also can easily cleaved cyclic ether to another yield where by using 1M of sulfuric acid can convert cyclic ether by 70% [17]. Other than sulfuric acid and hydrochloric acid, phosphoric acid are widely used especially in detergent products and agricultural fertilizers production because it is relatively inexpensive, however, it is a week acid due to its disassociation constant, therefore, it cannot compete with sulfuric acid and hydrochloric acid.

In this work, the use wet scrubber method was used to remove ammonia, trimethylamine and diethyl ether. The aims of this research are to determine if sulfuric acid can effectively remove all three type of gases and to determine the optimize condition on concentration of sulfuric acid and contact time in removing all the gases.

2. METHODOLOGY

2.1 Experiment Design

The contact of gas with liquid solvent technique is use in scrubber equipment in the industry. The purpose of the equipment are also to clean the flue gas from their process or the recover the valuable component. In this research, small scrubber bottle was used to act as scrubber mechanism for the gas absorption to happen in order to treat the gases.

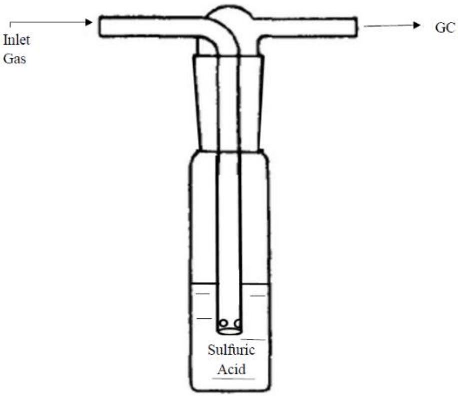


Figure 1: Experimental flow

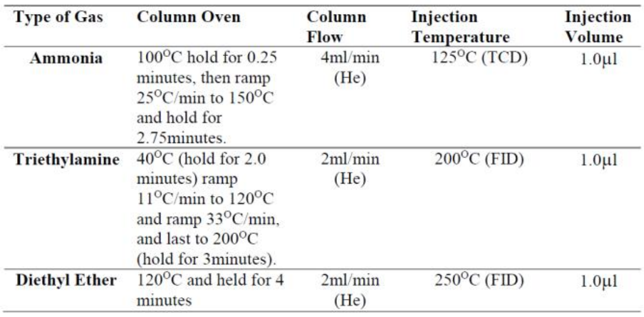
Figure 1 show the flow of procedure of the experiment. The gas will flow into the 250ml bottle and absorb by 25ml of sulfuric acid. The treated gas flow out the apparatus are collected and analyse using Gas Chromatography. All gases will have same method to treat them, but the method to produce the gas are different for each of them.

2.2 Analytical Method

All gases were tested using GC of ThermoFinnigan Trace GC type. The column used was Supelco Carboxen Fused Silica Capillary column with column length of 30m, internal diameter 0.53mm, and average thickness of 30 µm. The column inside the oven to ensure that the sample stay in gas form. [18]. All samples of gases were injected with injection volume of 1 µL. The data acquired was by using Chrom-Card data system version 2.3.1. Data of the equipment will shows the peak area of the gases in mvolt unit. Table 1 shows chromatographic condition for each type of gas [19][20].

Table 1: GC analysis for each type of gas

2.3 Methods for Specific Gas



Type of Gas	Column Oven	Column Flow	Injection Temperature	Injection Volume
Ammonia	100^{O}C hold for 0.25 minutes, then ramp 25^{O}C/min to 150^{O}C and hold for 2.75minutes.	4ml/min (He)	125^{O}C (TCD)	1.0µl
Triethylamine	40^{O}C (hold for 2.0 minutes) ramp 11^{O}C/min to 120^{O}C and ramp 33^{O}C/min, and last to 200^{O}C (hold for 3minutes).	2ml/min (He)	200^{O}C (FID)	1.0µl
Diethyl Ether	120^{O}C and held for 4 minutes	2ml/min (He)	250^{O}C (FID)	1.0µl

The methods given below are specified for each type of gas. As stated in experiment design, the procedure to treat the gas are the same, but the methods in producing the gases are different for each.

2.3.1 Ammonia

Ammonia gas was obtained from contact of ammonium chloride and sodium hydroxide in water. Ammonium chloride was purchased from Merck while sodium hydroxide was from Classic Chemical. 3g of ammonium chloride, 3g of sodium hydroxide and 2ml of distilled water. Reaction below shows the reaction of ammonium chloride and sodium hydroxide, which produce sodium chloride, ammonia gas and water.

$$NH_4Cl + NaOH \rightarrow NaCl + NH_3 + H_2O \tag{1}$$

3g of ammonium chloride and 3g of sodium hydroxide can produce 1.257L gas and the concentration is 61550 ppm. Ammonia gas that produce before and after the experiment were collected and injected into GC.

2.3.2 Triethylamine

Triethylamine solution was heated above boiling point, which is 130^{O}C. Triethylamine solution was purchased from Fisher Chemical with purity of 99%. The solution concentration is 7.2M. 7.2M are equivalent to 733680 ppm.

2.3.3 Diethyl Ether

Diethyl ether is one of type of ether group compound and it high volatile flammable. Diethyl ether solution was purchased from Merck with 99.5% purity. Diethyl ether solution were indirectly heated at 30^{O}C. Indirectly heat means that diethyl solution were immersed in water which is heated using hot plate at 35^{O}C. However, using thermometer the exact water temperature is determine which at 30^{O}C.

This technique were used because diethyl ether is a volatile highly flammable organic liquid. This means that with direct heat from the hot plate, it could get caught on fire. Since boiling point of diethyl ether is 34.6^{O}C, therefore 30^{O}C should be enough to vaporize the solution. This technique was combine by supplying nitrogen gas in the solution. In one hour, 45ml of diethyl ether were vaporise.

2.4 Sulfuric Acid

Sulfuric acid in this experiment was used as a solvent to treat the gases with gas absorption method. The solution was purchased from R&M chemical with 95% of purity and was diluted to 0.01M, 0.05M, 0.1M and 0.5M. The concentration used was not the same to treat all gases. The gases were treat with different concentration of sulfuric acid with different time taken. Table 2 shows the parameter on gases with concentration of sulfuric acid towards time.

Table 2: Parameter to treating the gas emission.

Type of Gas	Concentration of H_2SO_4(M)	Time (minutes)
Ammonia	0.05	5,10,15,20
	0.1	
	0.5	
Triethylamine	0.01	10, 15, 20
	0.05	
	0.1	
	0.5	
Diethyl Ether	0.01	5,10,15,20
	0.05	
	0.1	
	0.5	

3 RESULTS AND DISCUSSION

3.1 *Removal of Ammonia*

The collection of pure ammonia from the reaction and the gas was inject to the GC in order to obtain ammonia at time 0s. The experiment started by using 0.5M of sulfuric acid as solvent. The gas were pass through for 15 minutes and collected. The gas collected show no result in GC. Then, the experiment proceed with 5 minutes and 10 minutes but the result is still the same. 0.1M and 0.05M of sulfuric were used to capture ammonia gas and it shows increasing of percent removal.

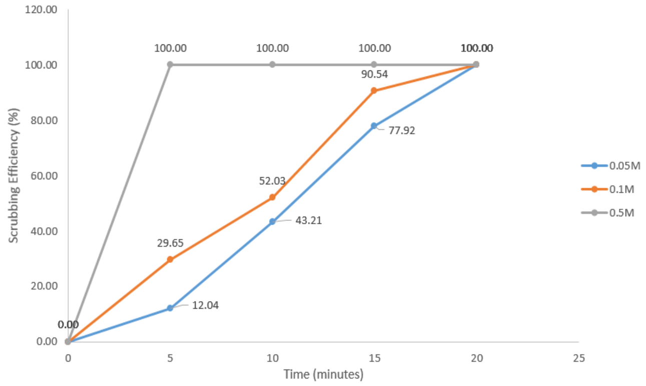


Figure 2: Ammonia removal at different concentration and time of sulfuric acid.

Figure 2 shows 0.5M is the most effective followed by 0.1M and 0.05M. It can be seen that 0.05M has the lowest efficiency. The lowest percent reduction of the ammonia is for 5 minutes of 0.05M sulfuric acid which it only removed 12%. As the time increase, the percent removal also increase to 43% and 77% for 10 and 15 minutes respectively. 0.5M of sulfuric acid is the most effective because it could remove ammonia for 100% even at 5 minutes contact time. The removal of ammonia using sulfuric acid occur due to the reaction which will produce $(NH_4)_2SO_4$ where it can be commercialized as ammonium sulfate soil fertilizer [21] [22].

Percent removal for 5 minute of 0.1M are higher than 0.05M with 29.65% removal. The same result shows for 10 minutes where 0.1M have 52.03% removal and 0.05M have removal of 43.21%. At 15 minutes, 0.1M has already meet 90.54% removal compared to 0.01M are only 77.92%. 100% removal can only be achieved at 20 minutes for both concentration. The sulfuric acid able to remove ammonia at any concentration and time. The similarities effectiveness of using acid in removal also showed in several studies [21] [23] [24]. Ammonia removal using waster were less efficient than acid where, using reverse osmosis water are only 85.4% removal with time 210 minutes while tap water only shows 64.6% removal [9]. The advantage of sulfuric acid documented in [24], where a test of removing ammonia with presence of carbon dioxide and it is found that sulfuric acid did not absorb any amount of carbon dioxide. This is important since several hundred parts per million of ammonia are normally present in air.

3.2 *Removal of Triethylamine*

The experiment was started with the biggest concentration of sulfuric acid with the longest time taken, however, 0.5M shows the removal of 100% for all time. Then the experiment proceed to 0.1M and it still shows the same result. Lowering the sulfuric acid concentration shows there are effluent of triethylamine gas from the scrubber bottle. 0.05M and 0.01M of sulfuric were used to capture triethylamine gas and it shows increasing of percent removal.

Figure 3: Triethylamine removal at different concentration and time of sulfuric acid.

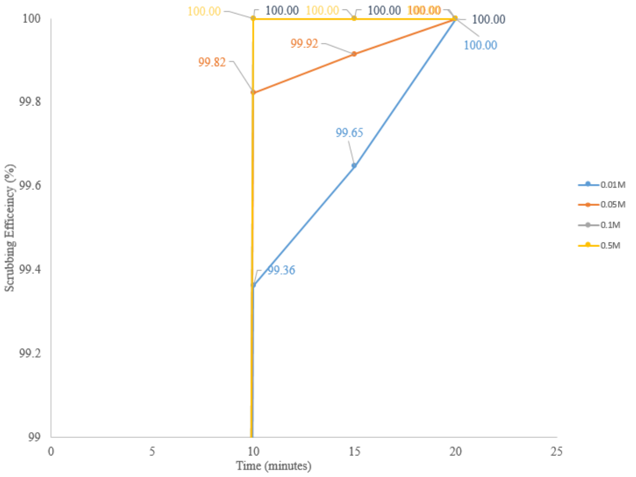


Figure 3 shows the percent removal for trimethylamine with different concentration of sulfuric acid towards reaction time. It can be observed that increasing of concentration with respect of time will increase scrubbing efficiency. Concentration of 0.05M and 0.01M were already achieved 99% reduction for 10 minutes. 0.01M could remove 99.36% at 10 minutes, 99.65% at 15 minutes, while, 0.05M could remove 99.82% at 10 minutes and 99.92% at 15 minutes. Even though the percent removal are close to each other but it still shows increment of percent removal. Both can completely remove amine for 20 minutes contact time. The reaction of amine and sulfuric acid will produce amine sulfate.

$$2[R_3N] + [H_2SO_4] \Rightarrow [R_3NH]_2[SO_4] \qquad (2)$$

Amine is base compound and can react with acid compound easily [25]. Sulfuric acid showed very effective in removing of triethylamine. The similarities result also showed in [11], where dimethyl amine can be removed by sulfuric acid with up to 95%. 0.01M of sulfuric acid are enough to remove completely 733,680 ppm of triethylamine for 20 minutes. Using lower concentration are economic since lower acidity solutions would reduce cost, corrosion and maintenance [9]. Pure triethylamine concentration was larger compared to ammonia, but the removal efficiency of triethylamine was higher. This is because, compared to ammonia, amine can reduce energy barrier of sulfuric acid more effectively and this is suggested by quantum chemical calculations [25].

3.3 *Removal of Diethyl Ether*

The collection of pure diethyl ether was inject to the GC in order to obtain diethyl ether at time 0s. The experiment started with the largest concentration of sulfuric acid with the longest time. The data shows that for 0.5M and 0.1M the diethyl ether are completely remove by sulfuric acid for 5,10,15 and 20 minutes. Therefore, the concentration of sulfuric are lowered to 0.05M and 0.01M.

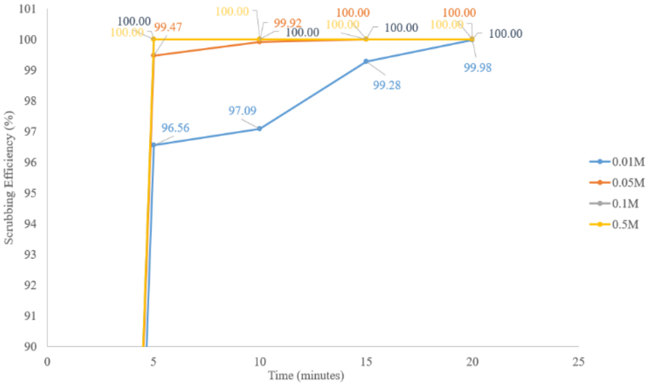


Figure 4: Diethyl Ether removal at different concentration and time of sulfuric acid.

Figure 4 shows diethyl ether removal using sulfuric acid solution at different concentration and contact time. The graph shows that 0.01M are less effective compared to other since the curve are below than other. For 0.05M of sulfuric acid, time contact of 5 minutes removes at 99.47% while for 10 minutes the scrubbing efficiency is 99.92%. It completely removes diethyl ether at 20 minutes. This is different with 0.01M, where at 20 minutes it can only remove 99.98%. It achieve 99% removal at 15 minutes while 0.05M obtain 99% removal at 5 minutes. 0.1M and 0.5M are able to completely remove all diethyl ether at 5 minutes makes it the most effectives.

The study shows that sulfuric acid are able to remove diethyl ether. The same outcome shows in [17], sulfuric acid can easily cleaved cyclic ether to another yield where by using 1M of sulfuric acid can convert cyclic ether by 70%. This conclude that sulfuric can remove diethyl ether from lowest concentration to the highest with shortest time contact and the removal efficiency continue to increase as the time contact increase.

4 CONCLUSION

In this study, removal process of the gases by sulfuric acid has been chosen. Acid are the most common compound used to neutralize based. The study shows that sulfuric acid are able to remove all three type of the base gases. It shows increasing of percent removal when in contact with sulfuric acid. The outcome display that, as increasing in time and concentration, the percentage removal of the bas gases are also increase.

0.5M concentration of sulfuric acid are most effective in removing all the three type of gases with 100% efficiency. The smallest amount removal for ammonia is at 0.05M of sulfuric at 5 minutes, while triethylamine is at 0.01M at 10 minutes and diethyl ether at 0.01M at 5 minutes. The optimum conditions are at the lowest and the shortest time taken. For ammonia, 0.5M of sulfuric acid at 5 minutes will achieved 100% efficiency removal. 100% efficiency removal of triethylamine achieved at 0.1M for 5 minutes. Lastly, the best condition for diethyl ether is 0.1M of sulfuric for 5 minutes of contact time. It can be conclude sulfuric acid are able to remove all type of base at different concentration and time.

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