**Regeneration of FGD Waste Liquors:
Production of Ammonium and Potassium
Sulfate Mixed Fertilizer**

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Progress Summary

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Confirmation of the feasibility of the K_2SO_4 process was made where a metathetical reaction was performed by adding potassium sulfate to the scrubbing liquor and the crystals formed as a result of the reaction were hydrolyzed to produce ammonium sulfate. Experimental investigations continued on precipitation and hydrolysis characteristics of the N-S compounds in the waste scrubbing liquor.

An overall test run was performed to demonstrate production of pure ammonium sulfate crystals by hydrolyzing precipitated potassium salts of N-S compounds.

To study crystallization characteristics of the N-S compounds, a bench scale 5 liter crystallizer was built. Preliminary operation showed that compared to a typical Mixed-Suspension-Mixed-Product-Removal (MSMPR) crystallizer, a Double-Draw-Off (DDO) crystallizer was superior by increasing the average size of the precipitated crystals of N-S compounds from 173 μm to 218 μm , thus enhancing their filterability.

For a fundamental improvement of the K_2SO_4 process, a different route of the process using different types of salts for precipitation of N-S compounds was investigated. A preliminary test showed that addition of lime/limestone (thioscorbic Mg-containing dolomite was O.K.) in a narrow pH range would precipitate N-S compounds in the scrubbing liquor. Detailed investigation needs to continue with emphasis on this lime/limestone process because it seems a better alternative process to the K_2SO_4 process in terms of economy and environmental safety. The K_2SO_4 process was originally proposed "in the spirit that it may evolve as experimental work progresses or suggestions are made for improvements".

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1. Introduction

1.1 Background of Project Process

A characteristic aspect of the Fe-EDTA wet scrubbing processes for simultaneous removal of SO_x and NO_x is accumulation of N-S compounds in the scrubbing liquor (Chang, 1986; Sloss *et al*, 1992). Therefore, regeneration of the scrubbing liquors involves removing the accumulated N-S compounds. This research highlights developments of the process which removes the N-S compounds and produces ammonium sulfate.

Processes developed to date can be classified into two types: Type I and II (Faucett, Maxwell, & Burnett, 1977; Martin, 1981). Type I processes are characterized as processes where the entire bleed stream from the scrubber is treated with sulfuric acid to decompose the N-S compounds by hydrolysis (e.g., Chisso Engineering Process and Mitsubishi Heavy Industry Process). Because sulfuric acid is added to the whole scrubbing liquor (which is mostly water, more than 95 wt %), the process requires a significant amount of sulfuric acid to maintain the level of acidity for hydrolysis as well as destroying the scrubbing liquor completely. Therefore, this type of process is not economical compared to a process which selectively separates N-S compounds from the scrubbing liquor and performs further treatment only on the separated compounds. Type II processes are characterized as processes where N-S compounds are selectively precipitated and decomposed to yield N_2 (e.g., Asahi Chemical Process). This type of process is less economical than our proposed process since no marketable byproduct is obtained after the decomposition of the N-S compounds.

The process which has been investigated with this year's DOE grant, possesses an advantage over the above-mentioned processes. N-S compounds in scrubbing liquors are first separated out after converting them to potassium salts by a metathetical reaction with

potassium sulfate (Chang, 1980, 1986). The introduction of the potassium sulfate originates from the fact that the potassium salts of N-S compounds are known to have relatively low solubility in an aqueous solution (Brauer, 1963). The separated potassium salts of the N-S compounds are then hydrolyzed under a sulfuric acid environment to produce a mixture of ammonium and potassium sulfate, which is salable as a mixed fertilizer. In the first quarterly report, the feasibility of this process was demonstrated. However, this process may be uneconomical since it requires purchasing potassium sulfate. Therefore, development of a different route process which uses salts other than potassium sulfate is of great concern.

The ultimate goal of this project is developing a regeneration process for waste scrubbing liquors through which N-S compounds in the scrubbing liquor are removed and then converted to produce a marketable byproduct. Because little information is available on N-S compound characteristics related to a regeneration process for waste scrubbing liquors, study on the originally proposed K_2SO_4 process was performed *"in the spirit that it may evolve as experimental work progresses or suggestions are made for improvements"*.

1.2 Chemistry of N-S Compounds in FGD Scrubbing Liquor

Knowledge of properties and reaction pathways of N-S compounds will be helpful to understanding and determining the regeneration process. Chang (1986) summarized reaction pathways of N-S compounds in metal chelate wet scrubbing as shown in Fig.1. Previous studies showed that the major N-S compounds in scrubbing liquors are hydroxylamine disulfonic acid (HADS), hydroxylamine monosulfonic acid (HAMS), amine trisulfonic acid (ATS), amine disulfonic acid (ADS), sulfamic acid (SA), and hydroxylamine (HA). At this point, however, it is not clear whether all of these N-S compounds can be removed from the scrubbing liquors as potassium salt precipitates since

Fig.1 Summary of reaction pathways of N-S compounds (Chang, 1986)

typical scrubbing concentrations of these N-S compounds are small (less than 100 mMol/liter). When based upon reports on major byproducts of several processes, the most probable precipitate will be potassium amine disulfonate (KADS). KADS is reported to possess low solubility in aqueous solution. Although KATS is more insoluble in aqueous solution than KADS, ATS is in general unstable in acidic environments and readily decomposes to ADS. Thus ADS seems to dominate ATS. Hydroxylamine sulfonic acids have high chances to react with abundant bisulfite ions to be converted to ADS. In addition, potassium salts of hydroxylamine sulfonic acids have higher solubilities than KADS. Therefore, KADS seems to be the major N-S compound in the potassium salt precipitate. This conjecture is made for preliminary data interpretation and comparison purpose in this report.

A point to note in the chemistry of N-S compounds is an expected reaction path which will be followed during hydrolysis of N-S compounds precipitated as potassium salts. From Fig.1, the expected reaction paths are summarized as follows.

ATS is hydrolyzed to ADS. As mentioned before, this reaction is relatively fast (Sisler & Audrieth, 1938; Nickless, 1968). ADS is hydrolyzed to yield sulphamate (Doyle & Davidson, 1949). This step is reported to be relatively slow. Sulphamate is converted by hydrolysis to ammonium and sulfate ions. This hydrolysis is reported to be very slow and seems to be a rate determining step in the process of hydrolysis from ATS for formation of ammonium ion (Maron & Berens, 1950; Candlin & Wilkins, 1960). From the path of hydrolysis, it seems difficult to describe overall hydrolysis in terms of reagents since in practice various N-S compounds of different concentration would be involved in

hydrolysis accompanying various side reactions. Fortunately, ammonium ion is an end product which is purely formed from decomposition of the N-S compounds by hydrolysis. Thus concentration of the ammonium ion in a hydrolyzed crop can be used as a measure of hydrolysis. Before hydrolysis, no ammonium ion will be present in a solution of N-S compounds precipitated from the scrubbing liquor. As hydrolysis progresses, the concentration of ammonium ion will increase and reach a saturation at complete conversion of the N-S compounds.

2. Experimental Studies

2.1 K_2SO_4 Process

In K_2SO_4 process, N-S compounds in scrubbing liquors are first separated out after converting them to potassium salts by a metathetical reaction with potassium sulfate (Chang, 1980, 1986). The separated potassium salts of the N-S compounds are then hydrolyzed under a sulfuric acid environment to produce a mixture of ammonium and potassium sulfate. Following the feasibility demonstration in the first quarter, experimental investigation on optimum process conditions continued with a set up of a 5-liter bench scale continuous crystallizer.

2.1.1 Precipitation of N-S Compounds in Continuous Crystallizers

In order to characterize precipitation of N-S compound, various properties of scrubbing liquor were determined and presented in Table.1. Among various chemical elements of scrubbing liquor, nitrogen is of our concern since the element would be a representative for content of the N-S compounds in the scrubbing liquor. The total nitrogen content in the scrubbing liquor was easily determined by elemental analysis as 0.67 Mol/liter of scrubbing liquor (or 0.84 wt % of the scrubbing liquor). Unfortunately, the

total nitrogen content by elemental analysis was found to include the contribution of EDTA ($C_{10}H_{16}N_2O_8$) and therefore was greater than the actual nitrogen content by N-S compounds. In order to describe precipitation characteristics of N-S compound, one needs to introduce a quantity and its measurement method for only N-S compounds in the scrubbing liquor. Hydrolyzable nitrogen content is determined by measuring the ammonium ion concentration with an ammonium ion selective electrode after completely hydrolyzing the mother liquor in a sulfuric acid environment. Since EDTA was observed not to be hydrolyzable under the same acidic condition, the hydrolyzable nitrogen content is a pure contribution of N-S compounds in the scrubbing liquor. The hydrolyzable nitrogen content in the scrubbing liquor was measured as 0.2 M/liter, which is smaller than the total nitrogen content. The concept of the hydrolyzable nitrogen content will be used next time for quantitatively describing the precipitation of the N-S compounds in the scrubbing liquor.

Other physical properties of the scrubbing liquor such as density and viscosity were measured to be similar to those of pure water.

Table 1 Various properties of Dravo scrubbing liquor.

Properties	
Total Nitrogen	0.67 M/liter
Hydrolyzable Nitrogen	0.20 M/liter
Density	1.11 g/cm ³
Viscosity	1.48 cP

In precipitation of N-S compounds, a process condition of primary concern is crystallizer operation which leads to increasing the size of the precipitates and thus enhancing their processibility. For this investigation, we set up a 5-liter bench scale

continuous crystallizer and compared characteristics of a MSMPR (Mixed Suspension Mixed Product Removal) and a DDO (Double Draw Off) crystallizer. The DDO crystallizer employs the principle of size-dependent residence time in order to improve the mean particle size of the product crystals. The DDO crystallizer is particularly suitable for application in low natural slurry density systems and its efficacy has been demonstrated in the precipitation of gypsum in wet FGD processes (Chang & Brna, 1984; Randolph *et al.*, 1984, 1990). Among the major advantages of the DDO configuration are the large particle size, high suspension density and low vessel fouling. A schematic diagram of the MSMPR and the DDO crystallizer are shown in Figs.2 (a) and (b), respectively. In DDO configuration shown in Fig.2(b), an overflow stream, Q_o , removes fine crystals at a classification cut size, L_F , drawn at a rate faster than the mixed under flow Q_u . This faster removal of fines allows the remaining particles to grow larger and thus if the cut size is small enough, the overall mean size is larger than that of the MSMPR. A detailed concept and theory of DDO operation is available in the literature (Randolph & Larsdon, 1988; White & Randolph, 1989).

In order to obtain an insight into effects of crystallizer type for precipitation of the N-S compounds, preliminary operation of MSMPR and DDO crystallizers was performed under the same condition. Fig.3 is a cumulative weight fraction vs. particle size plot for crystals precipitated by addition of K_2SO_4 in a MSMPR crystallizer (residence time, $\tau = 1.5$ hrs, K_2SO_4 feed rate = 7.2 g/100 ml liquor). Crystals obtained in the MSMPR crystallizer were shown evenly distributed in the size range. Weight average size of the crystals was 173 μm . Fig.4 is a cumulative weight fraction vs. particle size plot in a DDO crystallizer [residence time, $\tau = 1.5$ hrs; K_2SO_4 feed rate = 7.2 g/100 ml liquor; DDO ratio, $R = (Q_o + Q_u)/Q_u = 10$]. As operation of the crystallizer approached steady state with time, the fraction of crystals smaller than 200 μm gradually decreased while that of larger crystals increased. Weight average size of the crystals was measured as 228 μm , which is

(a)

(b)

Fig.2 Schematics of MSMPR (a) and DDO (b) crystallizer configurations. A MSMPR crystallizer is a constant stirred tank crystallizer. Crystal products in a MSMPR are typically distributed over a size range. In a DDO crystallizer, fine crystals are quickly removed as overflow and therefore more of larger crystals is found in the overall product. G = crystal growth rate; τ = residence time; R = DDO ratio defined as $(Q_0 + Q_u)/Q_u$.

Fig.3 Cumulative weight fraction vs. particle size plot for the crystals precipitated by K_2SO_4 in a 5 liter MSMPR crystallizer of $\tau = 1.5$ hrs and K_2SO_4 feed rate = 7.2 g/100 ml liquor. The crystals are evenly distributed over a size range. Weight average crystal size was 173 μm .

Fig.4 Cumulative weight fraction vs. particle size plot for the crystals precipitated by K_2SO_4 in a 5 liter DDO crystallizer of $\tau = 1.5$ hrs, $R = 10$, and K_2SO_4 feed rate = 7.2 g/100 ml liquor. As operation of the crystallizer approached steady state with time, the fraction of crystals smaller than 200 μm was shown to gradually decrease. Weight average crystal size was measured as 218 μm .

Fig.5 Photomicrographs for the crystals precipitated in the MSMPR (a) and DDO (b) crystallizer. Morphology of the crystals from different crystallizers are nearly similar but crystals formed in the MSMPR appeared relatively smaller in size.

approximately 50 μm bigger than that of the MSMPR crystallizer. This result tells one can obtain a larger crystal size by using a DDO crystallizer.

Figs.5 (a) and (b) are photomicrographs of the crystals obtained by MSMPR and DDO crystallizers. The morphology of the crystals from the two crystallizers are shown to be similar and composed of fine polycrystallites. Their morphology was similar to that of the crystals obtained using a batch crystallizer in the first quarter. However, one may see that the crystals precipitated in the MSMPR crystallizer are relatively smaller than those from the DDO crystallizer.

2.1.2 Hydrolysis and Production of Ammonium Sulfate

Hydrolysis characteristics were investigated for the crystals obtained from the MSMPR and DDO crystallizers and the results are also compared with that obtained in a batch crystallizer in the first quarter. Here, extent of hydrolysis is defined as the ratio of the existing ammonium ion concentration to total hydrolyzable nitrogen concentration since the goal of hydrolysis is converting the precipitated N-S compounds to ammonium sulfate regardless of formation of intermediate species. Therefore, the extent of hydrolysis is calculated as

$$\text{Extent of hydrolysis} = \frac{\text{Concentration of } \text{NH}_4^+ \text{ in the solution (Mol/liter)}}{\text{Concentration of } \text{NH}_4^+ \text{ at completion of hydrolysis (Mol/liter)}}$$

Concentration of ammonium ion was measured using an Orion ammonium ion selective electrode (Orion Model 95-12).

Fig.6 shows the extent of hydrolysis vs. time plot for the crystals obtained in three types of crystallizers. Crystals precipitated by a metathetical reaction with K_2SO_4 showed a

Fig.6 Extent of hydrolysis vs. time plot for the crystals obtained in three types of crystallizers. Crystals precipitated by a metathetical reaction with K_2SO_4 showed a similar trend of hydrolysis regardless of type of crystallizers. Compared to the crystals precipitated by K_2SO_4 , the crystals precipitated by addition of CaO were shown to be completely hydrolyzed in a shorter time. Hydrolysis was performed at $100^\circ C$ - $1M H_2SO_4$ acidic environment.

Fig.7 A typical photomicrograph of the crystals precipitated from the hydrolyzed crop after completion of hydrolysis. Since MgSO_4 was contained in the Dravo scrubbing liquor, the illustrated crystals are a mixture of ammonium sulfate, potassium sulfate, and magnesium sulfate.

similar trend of hydrolysis regardless of type of crystallizers. Fig.7 is a typical photomicrograph of the crystals precipitated from the hydrolyzed crop after completion of hydrolysis. Since MgSO_4 was contained in the Dravo scrubbing liquor, the illustrated crystals are a mixture of ammonium sulfate, potassium sulfate, and magnesium sulfate. It seemed difficult to identify individual salts because the salts form double salts.

2.2 An Alternative Process -- Lime/Limestone Process

In contrast to the K_2SO_4 process in which N-S compounds are precipitated as potassium salts by a metathetical reaction with K_2SO_4 , one may think about other types of processes which use different salts. Divers & Haga (1892) reported several metal salts of ADS such as Na, Ba, Sr, Ca, Hg, NH_4 , Ag, Mg, Zn, Ni, Co, Cu, Mn, Cd, etc.. Thus, in principle, sulfates of these metals may replace K_2SO_4 for precipitation of N-S compounds. A preliminary test in our lab showed that addition of lime/limestone (thioscorbic Mg-containing dolomite was O.K.) to scrubbing liquor in a narrow pH range would precipitate N-S compounds in the scrubbing liquor. An elemental analysis also showed that the precipitated crystals are probably mixture of CaSO_4 , CaSO_3 and CaADS (Calcium Amine Disulfonate). This lime/limestone process is a new process which seems better than the K_2SO_4 process because one does not need to purchase/introduce a new chemical additive to precipitate in the lime/limestone Fe-EDTA wet scrubbing processes.

Fig.8 is a typical photomicrograph of precipitated crystals by addition of lime to scrubbing liquor. Morphology of this crystal is in contrast with that obtained by K_2SO_4 addition (shown in Fig.5). Furthermore, these crystals were generally smaller in size. In comparing hydrolysis characteristics as shown in Fig.6, CaADS seemed to be completely hydrolyzed in a shorter time than KADS. This seems to result from chemical nature of

CaADS. In contrast to KADS, CaADS produces insoluble CaSO_4 by hydrolysis and thus hydrolysis of the CaADS become an irreversible reaction. This irreversible hydrolysis seems to enhance overall rate of CaADS conversion. Fig.9 is a typical micrograph of precipitates in a completely hydrolyzed crop. Similar to the crystals obtained by the K_2SO_4 process, these crystals seem to be a mixture of ammonium sulfate, calcium sulfate, and magnesium sulfate. Ammonium ion analysis of these crystals showed that ~ 42 wt % of the crystals precipitated by addition of lime to the scrubbing liquor was equivalent to CaADS. This result implies that the new lime/limestone process is as efficient in precipitating N-S compounds as the K_2SO_4 process which had ~ 50 wt % KADS equivalent of the crystals precipitated by adding 7.6 g K_2SO_4 /100 ml liquor.

3. Summary and Future Study

Precipitation and hydrolysis of the N-S compounds in the waste scrubbing liquor provided by Dravo Lime Co. was investigated. Precipitation of N-S compounds by a metatheical reaction with potassium sulfate was performed in continuous crystallizers. A preliminary operation showed that compared to a typical Mixed-Suspension-Mixed-Product-Removal (MSMPR) crystallizer, the Double-Draw-Off (DDO) crystallizer was superior by increasing the average size of the precipitated crystals of N-S compounds from 173 μm to 218 μm . However, the hydrolysis characteristics of the precipitated crystals were not dependent upon crystallizer type.

A brief description of a new process which uses lime/limestone for precipitation of N-S compounds in the scrubbing liquor is presented. Preliminary investigations showed the lime/limestone process is efficient in precipitating N-S compounds and the precipitated crystals were shown to be more easily hydrolyzed than potassium salts of N-S compounds. This lime/limestone process is a novel process which seems better than the K_2SO_4 process because one does not need to purchase/introduce a new chemical additive to precipitate in the lime/limestone Fe-EDTA wet scrubbing processes.

Up to the present, we focused on developing the K_2SO_4 process following our original proposal. However, the new lime/limestone process seems more advantageous in terms of economy and environmental safety. Therefore, it seems desirable changing research phase and putting an emphasis on the development of the lime/limestone process. Future study will include investigation of the DDO crystallizer operation to increase the size of precipitated crystals and thus to enhance their processibility. This study seems to be essential to the new lime/limestone process since the precipitated crystals are relatively small in size and thus poor in filterability.

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