A method of removing SO$_3$ from a flue gas stream includes providing a reaction compound selected from the group consisting of sodium carbonate, sodium bicarbonate, sodium sesquicarbonate, and mixtures thereof. The reaction compound is injected into the flue gas stream. The temperature of the flue gas is between about 500° F and about 850° F. The reaction compound is maintained in contact with the flue gas for a time sufficient to react a portion of the reaction compound with a portion of the SO$_3$ to reduce the concentration of the SO$_3$ in the flue gas stream.
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(71) Applicant (for all designated States except US): SOLVAY CHEMICALS, INC. [US/US]; 3333 Richmond Avenue, Houston, TX 77098-3009 (US).

(72) Inventor; and

(75) Inventor/Applicant (for US only): MAZIUK, Jonh, Jr [US/US]; 6502 Cobblestone Hill Street, Kingwood, TX 77345 (US).

(74) Agents: GILLIARD, Pierre et al.; Rue de Ransbeek, 310, B-1120 Bruxelles (BE).


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(54) Title: METHOD OF REMOVING SULFUR TRIOXIDE FROM A FLUE GAS STREAM

(57) Abstract: A method of removing SO3 from a flue gas stream includes providing a reaction compound selected from the group consisting of sodium carbonate, sodium bicarbonate, sodium sesquicarbonate, and mixtures thereof. The reaction compound is injected into the flue gas stream. The temperature of the flue gas is between about 500°F and about 850°F. The reaction compound is maintained in contact with the flue gas for a time sufficient to react a portion of the reaction compound with a portion of the SO3 to reduce the concentration of the SO3 in the flue gas stream.
Method of removing sulfur trioxide from a flue gas stream

The present invention relates to the purification of gases, and more particularly to a method of purifying flue gases which contain noxious gases such as SO₃.

SO₃ is a noxious gas that is produced from the combustion of sulfur-containing fuel. When present in flue gas, the SO₃ can form an acid mist that condenses in electrostatic precipitators, ducts or bag houses, causing corrosion. SO₃ at concentrations as low as 5-10 ppm in exhaust gas can also result in white, blue, purple, or black plumes from the cooling of the hot stack gas in the cooler air in the atmosphere.

The effort to reduce NOₓ emissions from coal-fired power plants via selective catalytic reactors (SCRs) has resulted in the unintended consequence of oxidizing SO₂ to SO₃ and thereby increasing total SO₃ emissions. SCRs employ a catalyst (typically vanadium pentoxide) to convert NOₓ to N₂ and H₂O with the addition of NH₃, but there is also an unintended oxidation of the SO₂ to SO₃.

Although the higher stack SO₃ concentrations are still relatively low, the emissions can sometimes produce a highly visible secondary plume, which, although unregulated, is nonetheless perceived by many to be problematic. Efforts to reduce the SO₃ levels to a point where no secondary SO₃ plume is visible can impede particulate collection for stations that employ electrostatic precipitators (ESPs). SO₃ in the flue gas absorbs onto the fly ash particles and lowers fly ash resistivity, thereby enabling the ESP to capture the particle by electrostatic means. Some plants actually inject SO₃ to lower fly ash resistivity when ash resistivity is too high.

SO₃ reacts with water vapor in the flue gas ducts of the coal power plant and forms vaporous H₂SO₄. A portion of this condenses out in the air heater baskets. Another portion of the sulfuric acid vapor can condense in the duct if the duct temperature is too low, thereby corroding the duct. The remaining acid vapor condenses either when the plume is quenched when it contacts the relatively cold atmosphere or when wet scrubbers are employed for flue gas desulfurization (FGD), in the scrubber’s quench zone. The rapid quenching of the acid vapor in the FGD tower results in a fine acid mist. The droplets are often too fine to be absorbed in the FGD tower or to be captured in the mist.
eliminator. Thus, there is only limited SO$_3$ removal by the FGD towers. If the sulfuric acid levels emitted from the stack are high enough, a secondary plume appears.

Dry sorbent injection (DSI) has been used with a variety of sorbents to remove SO$_3$ and other gases from flue gas. However, DSI has typically been done in the past at temperatures lower than around 370$^\circ$ F because equipment material, such as baghouse media, cannot withstand higher temperatures. Additionally, many sorbent materials sinter or melt at temperatures greater than around 400$^\circ$ F, which makes them less effective at removing gases. The reaction products of many sorbent materials also adhere to equipment and ducts, which requires frequent cleaning of the process equipment.

In one aspect, a method of removing SO$_3$ from a flue gas stream including SO$_3$ is provided. The method includes providing a reaction compound selected from the group consisting of sodium carbonate, sodium bicarbonate, sodium sesquicarbonate, and mixtures thereof. The reaction compound is injected into the flue gas stream. The temperature of the flue gas is between about 500$^\circ$ F and about 850$^\circ$ F. The reaction compound is maintained in contact with the flue gas for a time sufficient to react a portion of the reaction compound with a portion of the SO$_3$ to reduce the concentration of the SO$_3$ in the flue gas stream.

In another aspect, a method of removing SO$_3$ from a flue gas stream including at least about 3 ppm SO$_3$ includes providing a source of trona having a mean particle size between about 10 micron and about 40 micron. The trona is injected as a dry granular material into the flue gas stream. The temperature of the flue gas is between about 275$^\circ$ F and about 365$^\circ$ F. The trona is maintained in contact with the flue gas for a time sufficient to react a portion of the sodium sorbent with a portion of the SO$_3$ to reduce the concentration of the SO$_3$ in the flue gas stream. The reaction product comprises Na$_2$SO$_4$.

The foregoing paragraphs have been provided by way of general introduction, and are not intended to limit the scope of the following claims. The presently preferred embodiments, together with further advantages, will be best understood by reference to the following detailed description taken in conjunction with the accompanying drawings.

FIG. 1 is a phase diagram showing the reaction products of trona with SO$_3$ as a function of flue gas temperature and SO$_3$ concentration.

FIG. 2 is a schematic of one embodiment of a flue gas desulfurization system.
The invention is described with reference to the drawings in which like elements are referred to by like numerals. The relationship and functioning of the various elements of this invention are better understood by the following detailed description. However, the embodiments of this invention as described below are by way of example only, and the invention is not limited to the embodiments illustrated in the drawings.

Dry sorbent injection (DSI) has been used as a low cost alternative to a spray dry or wet scrubbing system for the removal of SO$_3$. In the DSI process, the sorbent is stored and injected dry into the flue duct where it reacts with the acid gas. Under certain processing conditions, the reaction product of the sorbent and the acid gas is a sticky ash. The sticky ash tends to stick to the process equipment and ducts, thus requiring frequent cleaning. Thus, it would be beneficial to have a process that minimizes the amount of sticky ash reaction product.

The present invention provides a method of removing SO$_3$ from a flue gas stream comprising SO$_3$ by injecting a reaction compound such as sodium sesquicarbonate, sodium bicarbonate, or soda ash into a flue gas stream to react with SO$_3$. Sodium sesquicarbonate is preferably provided from trona. Trona is a mineral that contains about 85-95% sodium sesquicarbonate (Na$_2$CO$_3$·NaHCO$_3$·2H$_2$O). A vast deposit of mineral trona is found in southwestern Wyoming near Green River. As used herein, the term “trona” includes other sources of sodium sesquicarbonate. The term “flue gas” includes the exhaust gas from any sort of combustion process (including coal, oil, natural gas, etc.). Flue gas typically includes acid gases such as SO$_2$, HCl, SO$_3$, and NO$_x$.

When heated at or above 275$^\circ$ F, sodium sesquicarbonate undergoes rapid calcination of contained sodium bicarbonate to sodium carbonate, as shown in the following reaction:

$$2 \text{[Na}_2\text{CO}_3 \cdot \text{NaHCO}_3 \cdot 2\text{H}_2\text{O}] \rightarrow 3\text{Na}_2\text{CO}_3 + 5\text{H}_2\text{O} + \text{CO}_2$$

Sodium bicarbonate undergoes a similar reaction at elevated temperatures:

$$2\text{NaHCO}_3 \rightarrow 3\text{Na}_2\text{CO}_3 + \text{H}_2\text{O} + \text{CO}_2$$

A preferred chemical reaction of the reaction compound with the SO$_3$ is represented below:

$$\text{Na}_2\text{CO}_3 + \text{SO}_3 \rightarrow \text{Na}_2\text{SO}_4 + \text{CO}_2$$

However, under certain conditions, undesirable reactions may occur which produce sodium bisulfate. If the sodium sesquicarbonate or sodium bicarbonate
is not completely calcined before reaction with SO$_3$, the following reaction occurs:

$$\text{NaHCO}_3 + \text{SO}_3 \rightarrow \text{NaHSO}_4 + \text{SO}_3$$

Under certain conditions, another undesirable reaction produces sodium bisulfate as represented below:

$$\text{Na}_2\text{CO}_3 + 2\text{SO}_3 + \text{H}_2\text{O} \rightarrow 2\text{NaHSO}_4 + \text{CO}_2$$

Sodium bisulfate is an acid salt with a low melt temperature and is unstable at high temperatures, decomposing as indicated in the following reaction:

$$2\text{NaHSO}_4 \rightarrow \text{Na}_2\text{S}_2\text{O}_7$$

The type of reaction product of the Na$_2$CO$_3$ and the SO$_3$ depends on the SO$_3$ concentration and the temperature of the flue gas. FIG. 1 is a phase diagram showing the typical reaction products of trona with SO$_3$ as a function of flue gas temperature and SO$_3$ concentration. In particular, above a certain SO$_3$ concentration, the reaction product can be solid NaHSO$_4$, liquid NaHSO$_4$, Na$_2$SO$_4$, or Na$_2$S$_2$O$_7$, depending on the flue gas temperature. The boundary between the liquid NaHSO$_4$ and the solid Na$_2$SO$_4$ at a temperature above 370° F may be represented by the equation log[SO$_3$]=0.009135T-2.456, where [SO$_3$] is the log base 10 of the SO$_3$ concentration in ppm and T is the flue gas temperature in °F. Liquid NaHSO$_4$ is particularly undesirable because it is “sticky” and tends to adhere to the process equipment, and cause other particulates, such as fly ash, to also stick to the equipment. Thus, it is desirable to operate the process under conditions where the amount of liquid NaHSO$_4$ reaction product is minimized. Thus, the process may be operated at a temperature below about 370° F, above about 525° F, or at a temperature and SO$_3$ concentration where

$$\text{log[SO}_3\text{]}<0.009135T-2.456.$$  

The temperature of the flue gas varies with the location in the injection system and may also vary somewhat with time during operation. As the temperature of the flue gas increases, the reaction product of the sodium compound and the SO$_3$ ranges from solid NaHSO$_4$, to liquid NaHSO$_4$, to solid Na$_2$SO$_4$ or Na$_2$S$_2$O$_7$. Therefore, to avoid the formation of sticky ash, the process is preferably operated in a suitable temperature range. In one embodiment, the temperature of the flue gas where the trona is injected is between about 500° F and about 850° F. The trona is maintained in contact with the flue gas for a time sufficient to react a portion of the trona with a portion of the SO$_3$ to reduce the concentration of the SO$_3$ in the flue gas stream. The temperature of the flue gas is preferably greater than about 500° F. The temperature of the flue gas is
preferably less than about 800° F, and most preferably less than about 750° F. The temperature of the flue gas is most preferably between about 525° F and about 750° F. In another embodiment, the temperature of the flue gas is between about 275° F and about 365° F. This temperature range is below the temperature for formation of the sticky NaHSO₄.

The SO₃ concentration of the flue gas stream to be treated is generally at least about 3 ppm, and commonly between about 10 ppm and about 200 ppm. In order to avoid the adhesion of waste material on the process equipment, when operated at flue gas temperatures greater than about 500° F the non-gaseous reaction product is preferably less than about 5% NaHSO₄, and most preferably less than about 1% NaHSO₄. The desired outlet SO₃ concentration of the gas stack is preferably less than about 50 ppm, more preferably less than about 20 ppm, even more preferably less than about 10 ppm, and most preferably less than about 5 ppm. The byproduct of the reaction is collected with fly ash.

Trona, like most alkali reagents, will tend to react more rapidly with the stronger acids in the gas stream first, and then after some residence time it will react with the weaker acids. Such gas constituents as HCl and SO₃ are strong acids and trona will react much more rapidly with these acids than it will with a weak acid such as SO₂. Thus, the injected reaction compound can be used to selectively remove SO₃ without substantially decreasing the amount of SO₂ in the flue gas stream.

A schematic of one embodiment of the process is shown in FIG. 2. The furnace or combustor 10 is fed with a fuel source 12, such as coal, and with air 14 to burn the fuel source 12. From the combustor 10, the combustion gases are conducted to a heat exchanger or air heater 30. Ambient air 32 may be injected to lower the flue gas temperature. A selective catalytic reduction (SCR) device 20 may be used to remove NOₓ gases. A bypass damper 22 can be opened to bypass the flue gas from the SCR. The outlet of the heat exchanger or air heater 30 is connected to a particulate collection device 50. The particulate collection device 50 removes particles made during the combustion process, such as fly ash, from the flue gas before it is conducted to an optional wet scrubber vessel 54 and then to the gas stack 60 for venting. The particulate collection device 50 may be an electrostatic precipitator (ESP). Other types of particulate collection devices, such as a baghouse, may also be used for solids removal. The baghouse contains filters for separating particles made during the combustion process from the flue gas.
The SO$_3$ removal system includes a source of reaction compound 40. The reaction compound is selected from sodium sesquicarbonate, sodium bicarbonate, and soda ash. The reaction compound is preferably provided as particles with a mean particle size between about 10 micron and about 40 micron, most preferably between about 24 micron and about 28 micron. The reaction compound is preferably in a dry granular form.

The reaction compound is preferably sodium sesquicarbonate in the form of trona. A suitable trona source is T-200® trona, which is a mechanically refined trona ore product available from Solvay Chemicals. T-200® trona contains about 97.5% sodium sesquicarbonate and has a mean particle size of about 24-28 micron. The SO$_3$ removal system may also include a ball mill pulverizer, or other type of mill, for decreasing and/or otherwise controlling the particle size of the trona or other reaction compound.

The reaction compound is conveyed from the reaction compound source 40 to the injector 42. The reaction compound may be conveyed pneumatically or by any other suitable method. Apparatus for injecting the reaction compound is schematically illustrated in FIG. 2. The injection apparatus 42 introduces the reaction compound into flue gas duct section 44, which is preferably disposed at a position upstream of the air heater 30. The injection system is preferably designed to maximize contact of the reaction compound with the SO$_3$ in the flue gas stream. Any type of injection apparatus known in the art may be used to introduce the reaction compound into the gas duct. For example, injection can be accomplished directly by a compressed air-driven eductor. Ambient air 32 may be injected to lower the flue gas temperature before the injection point 42.

The process requires no slurry equipment or reactor vessel if the reaction compound is stored and injected dry into the flue duct 44 where it reacts with the acid gas. However, the process may also be used with humidification of the flue gas or wet injection of the reaction compound. Additionally, the particulates can be collected wet through an existing wet scrubber vessel 54 should the process be used for trim scrubbing of acid mist. In particular, the flue gas desulfurization system may be operated so that the SO$_3$ removal is accomplished by injecting the reaction compound with the SO$_3$, while the majority of the SO$_2$ is removed by the wet scrubber 54.

The process may also be varied to control the flue gas temperature. For example, the flue gas temperature upstream of the trona may be adjusted to obtain the desired flue gas temperature where the reaction compound is injected.
Additionally, ambient air 32 may be introduced into the flue gas stream to lower
the flue gas temperature and the flue gas temperature monitored where the
reaction compound is injected. Other possible methods of controlling the flue
gas temperature include using heat exchanges and/or air coolers. The process
may also vary the trona injection location or include multiple locations for
reaction compound injection.

For the achievement of desulfurization, the reaction compound is
preferably injected at a rate with respect to the flow rate of the SO\textsubscript{3} to provide a
normalized stoichiometric ratio (NSR) of sodium to sulfur of about 1.0 or
greater. The NSR is a measure of the amount of reagent injected relative to the
amount theoretically required. The NSR expresses the stoichiometric amount of
sorbent required to react with all of the acid gas. For example, an NSR of 1.0
would mean that enough material was injected to theoretically yield 100 percent
removal of the SO\textsubscript{3} in the inlet flue gas; an NSR of 0.5 would theoretically yield
50 percent SO\textsubscript{3} removal. The reaction of SO\textsubscript{3} with the sodium carbonate is very
fast and efficient, so that a NSR of only one is generally required for SO\textsubscript{3}
removal. The reaction compound preferentially reacts with SO\textsubscript{3} over SO\textsubscript{2}, so
SO\textsubscript{3} will be removed even if large amounts of SO\textsubscript{2} are present. Preferably, an
NSR of less than 2.0 or more preferably less than 1.5 is used such that there is no
substantial reduction of the SO\textsubscript{2} concentration in the flue gas caused by reaction
with excess sorbent.

In one embodiment, the flue gas stream further comprises SO\textsubscript{2}, and
sufficient reaction compound is added to also remove some of the SO\textsubscript{2}. The
reaction compound is maintained in contact with the flue gas for a time sufficient
to react a portion of the reaction compound with a portion of the SO\textsubscript{2} to reduce
the concentration of the SO\textsubscript{2} in the flue gas stream. This may be particularly
useful in small plants, where it is more economical to have a single system for
removing both SO\textsubscript{2} and SO\textsubscript{3} rather than adding a wet scrubber to remove the
SO\textsubscript{3}.

Because NO\textsubscript{x} removal systems tend to oxidize existing SO\textsubscript{2} into SO\textsubscript{3}, the
injection system may also be combined with an NO\textsubscript{x} removal system. The trona
injection system may also be combined with other SO\textsubscript{x} removal systems, such as
sodium bicarbonate, lime, limestone, etc. in order to enhance performance or
remove additional hazardous gases such as HCl, NO\textsubscript{x}, and the like.

Surprisingly, it has been observed that when the temperature of the flue gaz
is between about 500°F and about 850°F (preferably between about 550°F and
about 750°F), or between about 275°F and about 365°F, the reaction product is not sticky. Solid build ups in the filter are avoided, in particular when it is a ESP. This effect is particularly pronounced in the upper temperature range.

Consequently, the invention concerns also the use of the method of removing SO₃ from a flue gas according to the invention and its preferred embodiments to avoid the formation of sticky reaction products.

**EXAMPLES**

Studies were conducted in an electric generation plant in Ohio using a hot side electrostatic precipitator (ESP) and no baghouse. The plant used a catalyst for NOₓ removal, which caused elevated SO₃ levels in the flue gas. The SO₃ concentration in the flue gas was between about 100 ppm and about 125 ppm. The trona used was T-200® from Solvay Chemicals.

**EXAMPLE 1**

T-200® trona was injected into the flue gas at a flue gas temperature of 367°F. A perforated plate of an ESP in the plant had significant solids buildup after operation of the SO₃ removal system for about two weeks.

**EXAMPLE 2**

The operation of Example 1 was repeated with the change that the trona was injected at a flue gas temperature below 365°F. In comparison to the perforated plate of Example 1, a perforated plate of an ESP in the plant had significantly less solids buildup after operation of the SO₃ removal system for two weeks than.

**EXAMPLE 3**

The operation of Example 1 is repeated with the change that the trona is injected into flue gas at a temperature of about 500°F. A perforated plate of an ESP in the plant is relatively free of solids buildup after operation of the SO₃ removal system for two weeks using T-200® trona.

The embodiments described above and shown herein are illustrative and not restrictive. The scope of the invention is indicated by the claims rather than by the foregoing description and attached drawings. The invention may be embodied in other specific forms without departing from the spirit of the invention. Accordingly, these and any other changes which come within the scope of the claims are intended to be embraced therein.
CLAIMS

1 - A method of removing SO₃ from a flue gas stream comprising SO₃, comprising:

- providing a reaction compound selected from the group consisting of sodium carbonate, sodium bicarbonate, sodium sesquicarbonate, and mixtures thereof;

- injecting the reaction compound into the flue gas stream, wherein the temperature of the flue gas is between about 500°F and about 850°F; and

- maintaining the reaction compound in contact with the flue gas for a time sufficient to react a portion of the reaction compound with a portion of the SO₃ to reduce the concentration of the SO₃ in the flue gas stream, wherein the reaction product of the reaction compound and the SO₃ is selected from the group consisting of Na₂SO₄, Na₂S₂O₇, and mixtures thereof.

2 - The method of claim 1 further comprising providing an NOₓ removal system upstream of the location where the reaction compound is injected.

3 - The method of claim 1 wherein the flue gas stream comprises at least about 3 ppm SO₃ upstream of the location where the reaction compound is injected.

4 - The method of claim 3 wherein the flue gas stream comprises between about 10 ppm and about 200 ppm SO₃ upstream of the location where the reaction compound is injected.

5 - The method of claim 1 wherein the reaction compound is sodium sesquicarbonate.

6 - The method of claim 1 wherein the reaction compound is provided in the form of particles with a mean particle size of less than about 40 micron.

7 - The method of claim 6 wherein the mean particle size of the reaction compound is between about 10 micron and about 40 micron.
8 - The method of claim 7 wherein the mean particle size of the reaction compound is between about 24 micron and about 28 micron.

9 - The method of claim 1 wherein the temperature of the flue gas is greater than about 550° F.

10 - The method of claim 1 wherein the temperature of the flue gas is less than about 750° F.

11 - The method of claim 1 wherein the temperature of the flue gas is between about 500° F and about 750° F.

12 - The method of claim 1 wherein the reaction compound is injected at a rate with respect to the flow rate of the SO$_3$ to provide a normalized stoichiometric ratio of sodium to sulfur of between about 1.0 and 1.5.

13 - The method of claim 1 wherein the reaction compound is injected as a dry material.

14 - The method of claim 1 further comprising milling the reaction compound to a desired mean particle size at a location proximate the flue gas stream.

15 - The method of claim 1 wherein the reaction product of the reaction compound and the SO$_3$ is selected from the group consisting of Na$_2$SO$_4$, Na$_2$S$_2$O$_7$, and mixtures thereof.

16 - The method of claim 1 wherein the flue gas stream further comprises SO$_3$, the method further comprising maintaining the reaction compound in contact with the flue gas for a time sufficient to react a portion of the reaction compound with a portion of the SO$_2$ to reduce the concentration of the SO$_2$ in the flue gas stream.

17 - A method of removing SO$_3$ from a flue gas stream, comprising:

- providing a source of trona;
- 11 -

- injecting the trona as a dry granular material into the flue gas stream, wherein the temperature of the flue gas is between about 500° F and about 850° F and wherein the flue gas stream comprises at least about 3 ppm SO₃; and

- maintaining the trona in contact with the flue gas for a time sufficient to react a portion of the trona with a portion of the SO₃ to reduce the concentration of the SO₃ in the flue gas stream.

18 - The method of claim 17 wherein the flue gas stream comprises between about 10 ppm and about 200 ppm SO₃ upstream of the location where the trona is injected.

19 - The method of claim 17 wherein the trona is provided in the form of particles with a mean particle size of between about 10 micron and about 40 micron.

20 - The method of claim 17 wherein the temperature of the flue gas is between about 500° F and about 750° F.

21 - The method of claim 17 wherein the reaction product of the reaction compound and the SO₃ is selected from the group consisting of Na₂SO₄, Na₂S₂O₇, and mixtures thereof.

22 - The method of claim 17 further comprising adjusting the flue gas temperature upstream of the trona to obtain the desired flue gas temperature where the trona is injected.

23 - The method of claim 17 wherein the adjusting further comprises introducing ambient air into the flue gas stream and monitoring the flue gas temperature where the trona is injected.

24 - The method of claim 17 wherein the adjusting further comprises controlling the flow of a material through a heat exchanger in communication with the flue gas.

25 - A method of removing SO₃ from a flue gas stream comprising at least about 3 ppm SO₃, comprising:
providing a source of trona having a mean particle size between about 10 micron and about 40 micron;

- introducing ambient air in the flue gas stream to reduce the temperature of the flue gas to less than about 365° F;

- injecting the trona as a dry granular material into the flue gas stream, wherein the temperature of the flue gas is between about 275° F and about 365° F; and

- maintaining the trona in contact with the flue gas for a time sufficient to react a portion of the sodium sorbent with a portion of the SO₃ to reduce the concentration of the SO₃ in the flue gas stream, wherein the reaction product is selected from solid phase Na₂SO₄, solid phase NaHSO₄, and mixtures thereof.

26 - The method of claim 25 wherein the flue gas stream comprises between about 10 ppm and about 200 ppm SO₃ upstream of the location where the trona is injected.