

Description

This invention relates to an improvement in liquid-gas contact apparatus and process where sulfur dioxide is absorbed into or reacted with the liquid. A particular application is in the absorber of the Wellman-Lord process for the removal of sulfur dioxide from exhaust gases by aqueous sulfite solution, particularly from combustion power plant flue gases, although it is also applicable to exhaust streams from smelters, sulfuric acid plants, or any other sulfur dioxide containing gas streams.

As an example of a process requiring an aqueous liquid-gas contact apparatus, the Wellman-Lord process of sulfur dioxide removal from stack gases employs an aqueous alkali sulfite solution (usually sodium sulfite) which chemically combines with the sulfur dioxide gas in an absorption tower by forming sodium bisulfite. The process includes a separate regeneration facility to convert the bisulfite back to sulfite and recover sulfur dioxide gas which is compressed and bottled or converted into sulfuric acid or to elemental sulfur. In the case of flue gases from coal fired plants, a separate unit for removal of fly ash and chlorides is included.

Because of large capital costs and high energy requirements to overcome the pressure drop in the absorber, in the regenerable Wellman-Lord process, non-regenerable flue gas desulfurization processes have been the more popular choice in the past, in spite of the major problem of disposal of solid waste produced by such processes.

The present invention is an improvement in absorption apparatus and its operation and in particular the process of the invention relates to the absorber of the Wellman-Lord process whereby capital costs and parasitic power requirements are reduced so as to make the regenerable process more economic and thus avoid the solid waste problem of the non-regenerable processes.

DESCRIPTION OF THE DRAWINGS

Figure 1 is a schematic of an adsorption column 10, indicating the position of a liquid distributor 11, an optional packing retainer 12, packing support 13, and conventional gas distributor and liquid collection, if necessary, 14, with the liquid and gas inlets and outlets indicated.

Figure 2 is an exploded broken view of a portion of a preferred liquid distributor having a supply trough 20, a spacing element 21, and a flow guide 22. Attached to flow guide 22 are drip fingers or rods 23 having ends adjacent packing (shown schematically) at 24.

Figure 3 is a schematic rib view, assembled, of the parts of a preferred distributor shown in Figure 2.

Figure 4 is an exploded schematic side view of a similar arrangement to Figure 2, but with the element 21 replaced by spacers 25.

Figure 5 is an enlarged cross-sectional view of a portion of element 21.

DESCRIPTION OF THE INVENTION

By employing liquid distribution at extremely low liquid flow rates and high efficiency random dumped packing, the present invention can achieve the absorption step in a single relatively short packed column of low pressure drop, and without the requirement of recycle of the absorbate solution prior to regeneration.

The flow rate for liquid fed to the top of the tower is from 0.000034 to 0.00068 m³/sec./m² (0.05 to 1.0 gallons per minute per square foot), or more preferably from 0.000136 to 0.000204 m³/sec./m² (0.2 to 0.3 gallons per minute per square foot). Only fresh absorbant solution is employed, without recycle. With the use of such low liquid flow rates, the tower packing is chosen so that its operation is compatible with the low liquid rate. One suitable type is that in which the operation is primarily one of combination and separation of droplets, rather than by a spreading of the liquid on large wetted areas of the packing. A suitable packing of this type is a high void volume packing shown in US-A-4 511 519 to Hsia, the disclosure of which is incorporated herein by reference. The packing has a large number of drip points, a relatively low surface area, but relatively long total length of non-aligned interacting edges.

Because of the low liquid rate of the present invention, with one half to one tenth of the liquid retention time of the conventional practice using trayed towers or a plurality of packed towers with trap trays, in the Wellman-Lord process, the problem of oxidation of sulfite to sulfate is reduced by an amount of 50% to as much as 90%. This is important in that the sulfate reduces the absorption efficiency of the liquid and thus must be removed in the regeneration process, adding capital and operational expenses to the process. In addition, the sulfate salts are a solid waste which must be disposed of in an environmentally safe manner.

In order to distribute liquid onto the packing so as to take advantage of the low flow rate a drip type distributor which provides at least 32.29 (3) and preferably at least 64.58 (6) to 96.87 (9) feed points

uniformly spaced per m² (square foot) may be employed. A suitable liquid distributor originally intended for non-polar liquids which can be adapted for this purpose is shown in US-A-4 264 538. While this type of distributor as disclosed in the patent was designed for organic liquids, it can be adapted for use in the present invention by use of hydrophilic coatings, as later described below.

Usually a metal (e.g. stainless steel) trough type distributor is used. The surfaces of the distributor, outside of the feed troughs which convey the liquid, should be coated with a hydrophilic microporous coating which causes a film of liquid to be formed on such surfaces as the liquid flows on them by gravity. The drip fingers, which are coated, should also all extend to be close to or contacting a surface of the packing (or packing retainer, if used). The distance should not be substantially greater than the diameter of the liquid droplets being fed, so as to avoid entrainment of the liquid by the countercurrent flowing gas. Such an arrangement produces maximum efficiency and can avoid the need for a demister at the exit of the absorber.

In the following examples a packing element as shown in Figures 7 through 10 of US-A-4 511 519 was employed, having a diameter of 88.9 mm. (3-1/2 inches) and an axial height of 31.75 mm, (1-1/4 inches). The packing depth was 2286 mm. (7.5 feet). A liquid distributor such as described in US-A-3 937 769 was employed, having 96.87 (9) drip fingers per m² (square foot). The operative surfaces of the distributor were coated with a resin latex drag resistant hydrophilic coating as described in US-A-4 467 070, sold by Hydromer, Inc., Whitehouse, N.J. The distributor also included a dimpled, perforated plate as shown at 21 in Figures 2, 3, and 5. The dimples were alternately facing in opposite directions. The dimples 50 of Fig. 5 in this case were 4.7625 mm (3/16 inches) apart, in an 0.2032 mm. (8 mil) steel sheet, to help distribute the liquid. The tower was 762 mm, (30 inches) in diameter. The liquid composition was composed as follows:

	Weight %
Na ₂ SO ₃	17.4
Na ₂ S ₂ O ₅	2.9
Na ₂ SO ₄	6.0
H ₂ O	73.7

The test results are shown in Table I.

Table II shows the pressure drop and the calculated mass transfer coefficient for each run. The highest mass transfer coefficients were obtained for the runs in which the gas rate was such that the tower was operating in the loading zone. That is, the gas rate was sufficiently high to cause increased liquid hold-up in the tower resulting in increased pressure drop and gas liquid contacting because of the added space taken up by the liquid.

Table I

Run No.	Liquid Rate m ³ /m ² (GPM/ft ²)	Gas Rate kg/s.m ² (Lbs/hr/ft ²)	Gas SO ₂ Parts/Million	
			Inlet	Outlet
1	0.0000985 (0.145)	2.4592 (1813.3)	717.2	160
2	0.0000964 (0.142)	4.5767 (3374.6)	711.7	220
3	0.0001066 (0.157)	4.7821 (3526.0)	944.5	510
4	0.0002119 (0.312)	4.9254 (3631.7)	1402.1	400
5	0.0001066 (0.157)	2.6638 (1964.1)	1065.7	600
6	0.0002200 (0.324)	4.7216 (3481.4)	676.0	80

Table II

Run No.	Pressure Drop kPa/m (Inches of H ₂ O/ft)	Mass Transfer Coefficient K _G a kg Moles/sec./m ³ kPa (LB Moles/hr-ft ³ -ATM)
1	0.0858 (0.105)	0.000588 (13.4)
2	0.3171 (0.388)	0.000970 (22.1)
3	0.3506 (0.429)	0.000681 (15.5)
4	0.3506 (0.429)	0.001238 (28.2)
5	0.0923 (0.113)	0.000299 (6.8)
6	0.3767 (0.461)	0.001730 (39.4)

The use of a single packed bed greatly reduces the capital cost of the absorber compared to present design practice used in Wellman-Lord plants. The single packed bed also greatly reduces the operating cost, primarily in the area of the blower horsepower needed to feed the gas through the absorber but also by eliminating the presently required liquid recycle pumps. The packed bed, to achieve these energy savings, preferably uses a high efficiency packing which can achieve in a minimum length of bed the necessary mass transfer contacting efficiency, and has sufficiently low pressure drop to make the energy savings possible.

Partial capital cost and energy saving benefits can be achieved by using less than optimum arrangements of this process invention. That is, rather than replacing all of the pump-around sections in prior absorption processes with a single packed bed, it is possible to combine some of the pump-around sections into two or three sections. These sections can still employ liquid recycle to achieve the high liquid rate operation but still have some pressure drop reduction. The energy savings here comes from the elimination of some of the trap trays. The elimination of the trap trays also saves considerable capital; the trap trays are very expensive because of their stainless steel construction.

One further partial simplified configuration is possible. This configuration uses two beds. The top section would be a packed section using the once through regenerated absorption solution at low liquid rate. Thus, making an off gas with the lowest possible SO₂ content because of low SO₂ vapor pressure in the regenerated solution. The lower bed would be recirculated for the benefit of the higher liquid rates for easier liquid distribution, and would tend to eliminate potential salt precipitation problems caused by dry areas.

Thus the single bed, with no pump around, employing low liquid rate may be used to absorb SO₂ from a gas stream directly from its source, or, as the final stage, from any intermediate pollution control apparatus including fly ash removal and chloride removal where required for coal burning power plants.

In normal operation typically a 10 to 20% stoichiometric excess of sorbent sulfite solution will be fed to the tower, based on the concentration of the sulfite and the liquid and gas feed rates. In some cases, however, it may be more desirable to fully react the sorbent liquid for efficient regeneration, than to remove all of the SO₂ from the gas. In such cases the liquid feed would be somewhat deficient, on a stoichiometric basis.

Claims

1. A method of removing SO₂ from a gas stream which comprises passing said gas stream to the bottom of at least one liquid-gas contact bed in a tower and feeding aqueous sulfite solution to the top of the bed, and withdrawing gas depleted in SO₂ from the top of the bed and bisulfite solution from the bottom of the bed, characterized by the fact that the SO₂ depletion is essentially completed in a single pass of the aqueous sulfite solution through the bed using an aqueous sulfite flow rate through the bed of from 0.000034 to 0.00068 m³/sec./m² and a trough type distributor having a hydrophilic surface for supplying a low liquid flow rate of the solution to the top of the bed, said bed containing random dumped packing for carrying out said liquid-gas contact.
2. A method according to claim 1, in which the flow rate is 0.000136 to 0.000204 m³/sec./m².
3. A method according to claim 1 or 2, in which the total sulfite content of the entering liquid per unit of time is at least equal to the sulfur dioxide content of the entering gas, per unit of time.
4. A method according to any one of the preceding claims, in which the aqueous sulfite is an alkali sulfite.



Table II

Run No.	Pressure Drop kPa/m (Inches of H ₂ O/ft)	Mass Transfer Coefficient K _{ga} kg Moles/sec./m ³ kPa (LB Moles/hr-ft ³ -ATM)
1	0.0858 (0.105)	0.000588 (13.4)
2	0.3171 (0.388)	0.000970 (22.1)
3	0.3506 (0.429)	0.000681 (15.5)
4	0.3506 (0.429)	0.001238 (28.2)
5	0.0923 (0.113)	0.000299 (6.8)
6	0.3767 (0.461)	0.001730 (39.4)

The use of a single packed bed greatly reduces the capital cost of the absorber compared to present design practice used in Wellman-Lord plants. The single packed bed also greatly reduces the operating cost, primarily in the area of the blower horsepower needed to feed the gas through the absorber but also by eliminating the presently required liquid recycle pumps. The packed bed, to achieve these energy savings, preferably uses a high efficiency packing which can achieve in a minimum length of bed the necessary mass transfer contacting efficiency, and has sufficiently low pressure drop to make the energy savings possible.

Partial capital cost and energy saving benefits can be achieved by using less than optimum arrangements of this process invention. That is, rather than replacing all of the pump-around sections in prior absorption processes with a single packed bed, it is possible to combine some of the pump-around sections into two or three sections. These sections can still employ liquid recycle to achieve the high liquid rate operation but still have some pressure drop reduction. The energy savings here comes from the elimination of some of the trap trays. The elimination of the trap trays also saves considerable capital; the trap trays are very expensive because of their stainless steel construction.

One further partial simplified configuration is possible. This configuration uses two beds. The top section would be a packed section using the once through regenerated absorption solution at low liquid rate. Thus, making an off gas with the lowest possible SO₂ content because of low SO₂ vapor pressure in the regenerated solution. The lower bed would be recirculated for the benefit of the higher liquid rates for easier liquid distribution, and would tend to eliminate potential salt precipitation problems caused by dry areas.

Thus the single bed, with no pump around, employing low liquid rate may be used to absorb SO₂ from a gas stream directly from its source, or, as the final stage, from any intermediate pollution control apparatus including fly ash removal and chloride removal where required for coal burning power plants.

In normal operation typically a 10 to 20% stoichiometric excess of sorbent sulfite solution will be fed to the tower, based on the concentration of the sulfite and the liquid and gas feed rates. In some cases, however, it may be more desirable to fully react the sorbent liquid for efficient regeneration, than to remove all of the SO₂ from the gas. In such cases the liquid feed would be somewhat deficient, on a stoichiometric basis.

Claims

1. A method of removing SO₂ from a gas stream which comprises passing said gas stream to the bottom of at least one liquid-gas contact bed in a tower and feeding aqueous sulfite solution to the top of the bed, and withdrawing gas depleted in SO₂ from the top of the bed and bisulfite solution from the bottom of the bed, characterized by the fact that the SO₂ depletion is essentially completed in a single pass of the aqueous sulfite solution through the bed using an aqueous sulfite flow rate through the bed of from 0.000034 to 0.00068 m³/sec./m² and a trough type distributor having a hydrophilic surface for supplying a low liquid flow rate of the solution to the top of the bed, said bed containing random dumped packing for carrying out said liquid-gas contact.
2. A method according to claim 1, in which the flow rate is 0.000136 to 0.000204 m³/sec./m².
3. A method according to claim 1 or 2, in which the total sulfite content of the entering liquid per unit of time is at least equal to the sulfur dioxide content of the entering gas, per unit of time.
4. A method according to any one of the preceding claims, in which the aqueous sulfite is an alkali sulfite.

5. A method according to any one of the preceding claims, in which the liquid is distributed from drip fingers having said hydrophilic surface.
6. A method according to claim 5, in which the drip fingers are constructed of stainless steel having an organic hydrophilic coated thereon.

Patentansprüche

1. Verfahren zur Entfernung von SO_2 aus einem Gasstrom, bei dem man den Gasstrom dem Boden zumindest eines Flüssigkeits-Gas-Kontaktbettes in einem Turm zuleitet und wäßrige Sulfittlösung dem oberen Ende des Bettes zuführt und an SO_2 verarmtes Gas von dem oberen Ende des Bettes und Bisulfittlösung von dem Boden des Bettes abzieht, dadurch gekennzeichnet, daß die SO_2 -Verarmung im wesentlichen in einem einzigen Durchgang der wäßrigen Sulfittlösung durch das Bett unter Verwendung einer Fließgeschwindigkeit des wäßrigen Sulfits durch das Bett von $0,000034$ bis $0,00068 \text{ m}^3/\text{sec}/\text{m}^2$ und eines Verteilers vom Wannentyp mit einer hydrophilen Oberfläche für die Schaffung einer niedrigen Flüssigkeitsfließgeschwindigkeit der Lösung zu dem oberen Ende des Bettes beendet wird, wobei das Bett eine random-geschüttete Packung für die Durchführung des Flüssigkeits-Gas-Kontaktes enthält.
2. Verfahren gemäß Anspruch 1, worin die Fließgeschwindigkeit $0,000136$ bis $0,000204 \text{ m}^3/\text{sec}/\text{m}^2$ beträgt.
3. Verfahren gemäß Anspruch 1 oder 2, worin der Gesamtsulfitgehalt der je Zeiteinheit eintretenden Flüssigkeit zumindest dem Schwefeldioxidgehalt des je Zeiteinheit eintretenden Gases entspricht.
4. Verfahren gemäß einem der vorhergehenden Ansprüche, worin das wäßrige Sulfid ein Alkalisulfid ist.
5. Verfahren gemäß einem der vorhergehenden Ansprüche, worin die Flüssigkeit von Tropffingern, die die hydrophile Oberfläche aufweisen, verteilt wird.
6. Verfahren gemäß Anspruch 5, worin die Tropffinger aus rostfreiem Stahl mit einem organischen hydrophilen Überzug hierauf konstruiert sind.

Revendications

1. Un procédé pour éliminer le dioxyde de soufre d'un courant de gaz, procédé selon lequel on fait passer ledit courant de gaz par le fond d'au moins un lit de contact liquide-gaz dans une tour et on alimente en solution aqueuse de sulfure par le sommet de la tour, et on prélève le gaz exempt de SO_2 par le sommet du lit et la solution de bisulfure par le fond du lit, caractérisé par le fait que l'élimination du dioxyde de soufre est essentiellement terminée en une simple passe de solution aqueuse de sulfure à travers le lit en utilisant un débit d'écoulement du sulfure aqueux à travers le lit de $0,000034$ à $0,00068 \text{ m}^3/\text{s}/\text{m}^2$ et un distributeur du type cuve présentant une surface hydrophile pour fournir un faible débit d'écoulement de liquide de la solution par le sommet du lit, ledit lit renfermant un garnissage disposé au hasard pour la réalisation dudit contact liquide-gaz.
2. Un procédé selon la revendication 1, dans lequel le débit d'écoulement est de $0,000136$ à $0,000204 \text{ m}^3/\text{s}/\text{m}^2$.
3. Un procédé selon la revendication 1 ou 2, dans lequel la teneur totale en sulfure du liquide entrant par unité de temps est au moins égale à la teneur en dioxyde de soufre du gaz pénétrant, par unité de temps.
4. Un procédé selon l'une quelconque des revendications précédentes, dans lequel le sulfure aqueux est un sulfure alcalin.
5. Un procédé selon l'une quelconque des revendications précédentes, dans lequel le liquide est distribué à partir de pattes de ruissellement présentant une surface hydrophile.



6. Un procédé selon la revendication 5, dans lequel les pattes de ruissellement sont réalisées en acier inoxydable présentant un revêtement hydrophile y appliqué.

5

10

15

20

25

30

35

45

50

55

