

Mass Transfer Theory – Practical Approach

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1 Introduction

This document was produced and published by Gary Slenders – Principal Process Engineer for AOMC Pty Ltd as part of the education series for Engineering Student in the field of Process, Chemical, and Environmental Engineers as a means of providing a better understanding of the fundamental processes involved in Odour Control. The information contained in this document is based on established material currently in the public domain. You are free to use this material, we ask only that you provide appropriate recognition to the author if you use a significant portion of this document for educational or commercial purposes.

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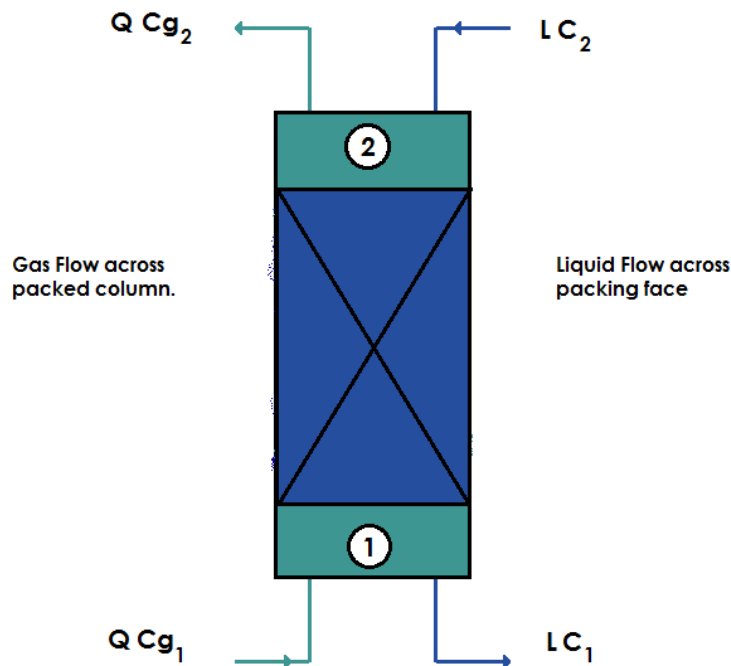
The key purpose of this document is to simplify the understanding of what Mass and Heat transfer is all about. As such the information presented is not as rigorous presentation as seen in the standard texts. For a more standardised assessment, we always recommend going back to the source documents.

This document is the third part of the series of documentation presented in relation to odour control. In systems, such as BioTrickling Filters and BioFilters the entire process of the removal of odour from the gas stream is based on advanced mass transfer concepts. Further to this is the removal of those odorous compounds through liquid phase extraction is based on advanced chemistry. The limit in this paper only extends to the consideration of inorganic chemistry, in later papers the subject of biochemistry is addressed.

As previously discussed in Introduction to Mass Transfer, the analogies that exist between the Mass and Heat transfer mechanisms suggest that it is important to understand both processes. At a later stage, we discuss processes involving both mass and heat transfer in examples such as pack column humidifiers.

2 Mass Transfer in the Standard Packed Column

The simplest form of operational mass transfer consists of a standing column with a level of random packing fixed into it. There is water flow from the top and air flow from the underside to the top.



The configuration is based on the factors that:

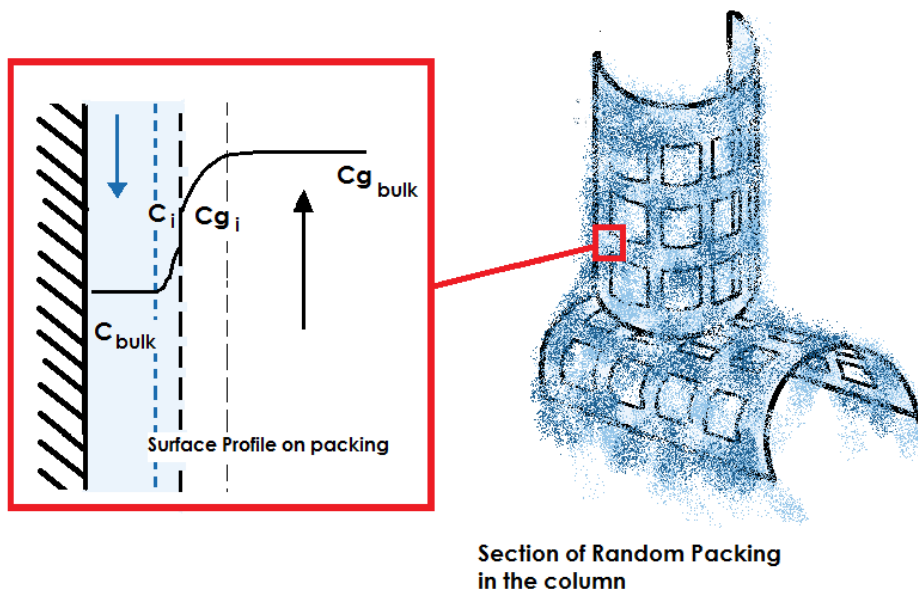
- The liquid phase is fully and evenly distributed across the entire surface of the random packing
- The gas phase is fully occupying the volume of the scrubber and evenly distributes the air flow across the full width of the column

The overall mass transfer represents the operating line on a graph of the gas phase vs liquid phase concentration

$$M' = Q(Cg_1 - Cg_2) = L(C_1 - C_2) \quad dM' = LdC \quad LdC - QdCg = 0$$

It is possible to determine the inlet and outlet conditions and at any point in the column given the linearity between the conditions through the column. Most prescribed texts including Faust and Perry's go to great length to suggest non-linearity as Q and L can change as Cg and C change, but in context of a functioning odour control systems this would never be the case because of the small concentrations involved.

The process of mass transfer is dependent on the interaction at the interface between the liquid and gas phases.



As with the analogy with heat transfer the rate of transfer is dependent on the driving force. The difference is that unlike temperature there is a barrier to overcome through the change of phase from gas to liquid and there needs to be an understanding of how that relationship works.

$$QdC_g = -k_G a(C_g - C_{g,i})dV \quad LdC = k_L a(C - C_i)dV \quad \text{and} \quad C_{g,i} = f(C_i)$$

There is a gradient profile of contaminant concentration on the liquid and gas phases. The profile exists only within the laminar sublayer just immediately at the interface surface. The interface contains an equilibrium between the liquid and gas phases in the immediate sublayer. That equilibrium is dependent on the concentration and temperature. For systems with dilute levels of contaminants this equilibrium is expressed as the Henry's Law constant for which

$$C_{g,i} = H C_i \quad \text{where} \quad H = H_0 e^{-k\left(\frac{1}{T} - \frac{1}{T_0}\right)}$$

Generally, Henry's Law constants are listed with standard reference to 20° C or 25° C and the temperature are stated as Absolute Temperatures (Kelvin)

3 Traditional design method

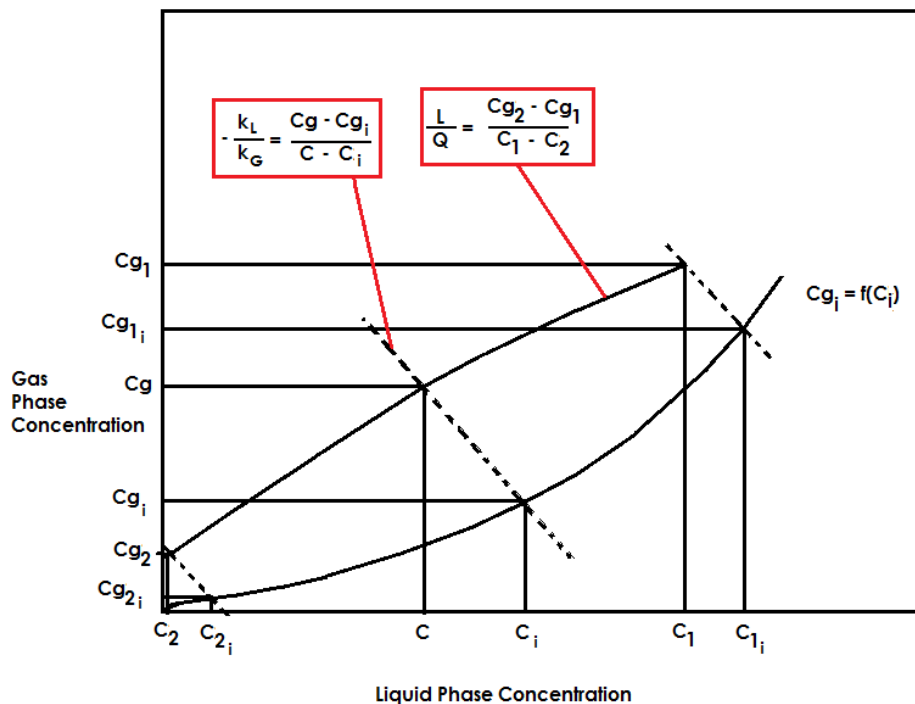
There is a traditional method for designing scrubber towers based on the above equations that is incredibly tedious. Based on fundamental principles it is satisfactory for example purposes, but thereafter should be ignored.

$$\frac{L}{Q} = \frac{(Cg_1 - Cg_2)}{(C_1 - C_2)} \quad Cg_i = f(C_i) \quad \text{and} \quad \frac{-k_L a}{k_G a} = \frac{(Cg - Cg_i)}{(C - C_i)}$$

Strictly speaking the operating curve formula is not represented correctly as there is an incremental change in the volume as contaminant concentration changes

$$\frac{L(1 + C)}{Q(1 + Cg)} = \frac{(Cg - Cg')}{(C - C')}$$

So even whatever is supposed to be linear is never assumed to be linear.



This is the commonly graphed operating and equilibrium curve that demonstrates the profile across a packed column.

The equation that allows the column packing volume to be determined is

$$V = \frac{Q}{-k_G a} \int_{C_{g1}}^{C_{g2}} \frac{dC_g}{(C_g - C_{g_i})} = \frac{Q}{-k_G a} \sum_{C_{g1}}^{C_{g2}} \frac{\Delta C_g}{(C_g - C_{g_i})}$$

The process going forward is to determine the column volume is to:

- Select an incremental value for C_g
- Starting at C_{g1} incrementing down, as C_g incrementing backwards, for every value of C_g find C , C_{g_i} , and C_i based around drawing a line of gradient $-k_G/k_L$ and find $C_g - C_{g_i}$
- Select the inverse value and continue repeating right down to C_{g2}
- After what can only be an eternity find the sum of the inverse $C_g - C_{g_i}$
- Calculate Q and $k_G a$ as some mysterious average across the column

The final product is the volume of packing. Note that as the mass transfer coefficient as determined from Chilton - Colburn analogy or the surface renewal theory or any other method requires details of the velocity through the column and therefore the diameter / cross sectional area is required to be determined and consequently the height of the column can be determined.

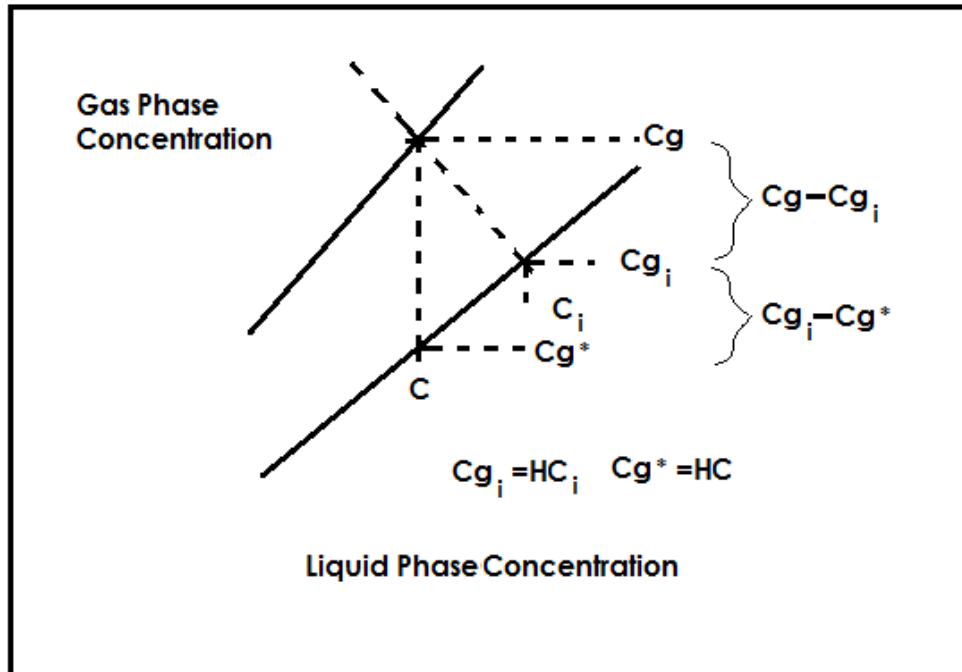
The above process works, but is not practical. This method is adopted for reasons that have no relevance to the type of systems we are considering:

- The movement of contaminants from the gas phase to the liquid phase represents a change in volume of 0.001% of total flow. Measurement error for air flow is 10%. Any consideration of the flow of contaminants from the gas phase to the liquid phase impacting on the overall gas flowrate only holds true when considering multistage distillation columns, but has no relevance to odour control.
- Up to a point of 2 – 5 % by volume 20000 – 50000 ppm there is a linearity between the equilibrium conditions in the gas phase and the equilibrium conditions in the liquid phase. The highest level of measure hydrogen sulphide in sewer network is only around 5000 ppm. Assuming a linear relationship between equilibrium concentration of the gas and liquid phase has no detrimental impact on design. Henry's Law should always be used.

Assuming linearity greatly simplifies the design and does not sacrifice in any way the accuracy of that design.

4 Practical method of column design

First consideration is whether there is a more simpler way of determining the equilibrium concentration based on the determined operating point rather than having to apply a linear intercept to an interface curve



The concept of the overall mass transfer coefficient, its analogy to overall heat transfer coefficient has some merit as again it is avoiding determining the interface conditions which is calculable but is time consuming. It is again the case that the bulk conditions of both the gas phase and liquid phase are the easiest to work with.

$$QdC_g = -K_G a(C_g - C_g^*)dV = -k_G a(C_g - C_{g_i})dV = -LdC = -k_L a(C - C_i)dV$$

$$-LdC = -\frac{k_L a}{H}(HC - HC_i)dV = \frac{k_L a}{H}(C_{g_i} - C_g^*)dV$$

$$(C_g - C_g^*) = (C_g - C_{g_i}) + (C_{g_i} - C_g^*)$$

$$\frac{-QdC_g}{K_G a dV} = \frac{-QdC_g}{k_G a dV} + \frac{-QdC_g}{\frac{k_L a}{H}}$$

This can be simplified down to:

$$\frac{1}{K_G} = \frac{1}{k_G} + \frac{H}{k_L} \text{ where } QdCg = -K_G a(Cg - Cg^*)dV$$

The next assumption is that the operating curve is not a curve but a straight line. Therefore, there is a linear relationship between Cg & C , Cg^* & C , therefore a linear relationship between Cg and Cg^*

$$(Cg - Cg^*) = \alpha + \beta Cg, (Cg_1 - Cg^*_1) = \alpha + \beta Cg_1, (Cg_2 - Cg^*_2) = \alpha + \beta Cg_2$$

$$\frac{Q}{-K_G a} \int_{Cg_1}^{Cg_2} \frac{dCg}{(Cg - Cg^*)} = \frac{Q}{-K_G a} \int_{Cg_1}^{Cg_2} \frac{dCg}{(\alpha + \beta Cg)} = \int_0^V dV$$

$$V = \frac{Q}{-K_G a \beta} \ln \left| \frac{\alpha + \beta Cg_2}{\alpha + \beta Cg_1} \right| = \frac{Q}{K_G a \beta} \ln \left| \frac{(Cg_1 - Cg^*_1)}{(Cg_2 - Cg^*_2)} \right|$$

$$(Cg_1 - Cg^*_1) - (Cg_2 - Cg^*_2) = \beta(Cg_1 - Cg_2)$$

$$\frac{1}{\beta} = \frac{(Cg_1 - Cg_2)}{(Cg_1 - Cg^*_1) - (Cg_2 - Cg^*_2)}$$

$$V = \frac{Q(Cg_1 - Cg_2)}{K_G a} \frac{\ln \left| \frac{(Cg_1 - Cg^*_1)}{(Cg_2 - Cg^*_2)} \right|}{(Cg_1 - Cg^*_1) - (Cg_2 - Cg^*_2)}$$

It is no coincidence that there is a clear analogy between linearity in mass transfer and heat transfer.

$$M' = Q(Cg_1 - Cg_2) = K_G a V \frac{(Cg_1 - Cg^*_1) - (Cg_2 - Cg^*_2)}{\ln \left| \frac{(Cg_1 - Cg^*_1)}{(Cg_2 - Cg^*_2)} \right|}$$

$$E = Q_H \rho_H C p_H (T_1 - T_2) = U A_o \frac{(T_1 - T'_1) - (T_2 - T'_2)}{\ln \left| \frac{(T_1 - T'_1)}{(T_2 - T'_2)} \right|}$$

$$V = \frac{Q(Cg_1 - Cg_2)}{K_G a} \frac{\ln \left[\frac{(Cg_1 - Cg^*_1)}{(Cg_2 - Cg^*_2)} \right]}{(Cg_1 - Cg^*_1) - (Cg_2 - Cg^*_2)}$$

This formula makes the calculation of the volume of packing simple.

This equation by which wet chemical scrubbers are able to be designed only requires:

- Finding the corresponding inlet and outlet gas and liquid phase conditions
- Finding the overall mass transfer coefficient.

5 Improving performance of the scrubber

This simplified formula provides a direction towards reducing the volume of packing required to achieve the mass transfer results.

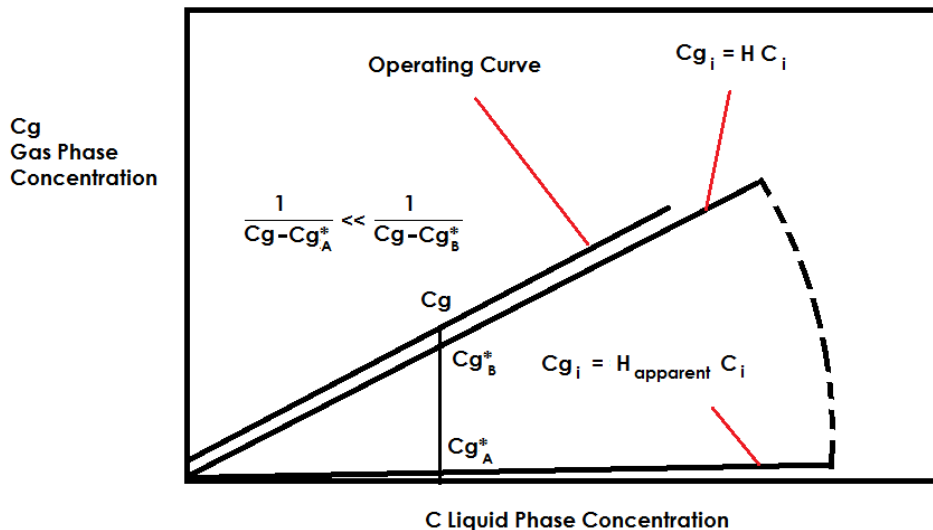
The parameters that can impact on performance are:

- Overall mass transfer coefficient – which has the only practical variable to change and that is the liquid and gas velocities within the column. All other parameters are attributes of air, water, and the contaminant. The limits are flooding and pressure drop across the bed.
- Specific surface area can be increased and is certainly a critical factor in design. There are practical limits that relate to pressure drop across the bed. Realistically a can only be increased to between 350 to 800 square meters per cubic meters
- Cg^* is a property of the contaminant but can be influenced using chemistry

The optimum value occurs when it is possible to drive C_{g1}^* and C_{g2}^* to a value approaching zero:

$$V = \frac{Q(Cg_1 - Cg_2)}{K_G a} \frac{\ln \left[\frac{(Cg_1 - 0)}{(Cg_2 - 0)} \right]}{(Cg_1 - 0) - (Cg_2 - 0)} = \frac{Q}{K_G a} \ln \left[\frac{Cg_1}{Cg_2} \right]$$

A method to reduce the interface liquid concentration of the contaminant to close to zero is required to significantly reduce the column volume and provide a practical design.



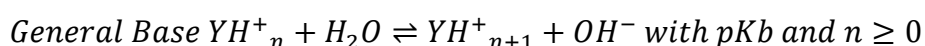
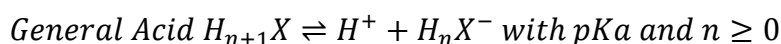
When looking at the application of scrubbing towers toward removing hydrogen sulphide in nearly all cases water alone is never used, and they operate with much greater efficiency and performance than if water alone is used. The selection of chemical is critical for the performance of the system.

6 Influence of pH on changes in chemical species concentration

The best way to consider how the introduction of chemicals influences the operations of scrubbers is to look at two specific examples. That is of ammonia in the presence of hydrochloric acid or sulphuric acid and hydrogen sulphide in the presence of caustic.

The chemistry of compounds such as Ammonia and Hydrogen Sulphide are classic Brønsted–Lowry acid–base which works on the principal of production of conjugate acid or bases in solution.

In general form, they are either release or attraction hydrogen ions under reversible conditions in water, referred to as disassociation:

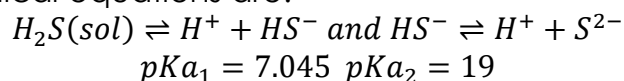


To which:

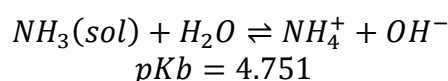
$$\frac{[H^+][H_nX^-]}{[H_{n+1}X]} = 10^{-pK_a} \text{ and } \frac{[YH_{n+1}^+][OH^-]}{[YH_n^+]} = 10^{-pK_b}$$

The square brackets represent activity of the ionic species, but again for simplicity it can be assumed to be the concentration of the ion species. The equilibrium equations determine the final composition of the solution. This differs to the added volume of acid or base by the reversing reaction to establish an equilibrium.

When specifically applied to Hydrogen Sulphide and to Ammonia, the dissociation chemical equations are:

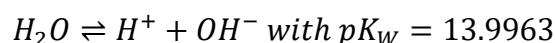


$$pK_{a1} = 7.045 \quad pK_{a2} = 19$$



$$pK_b = 4.751$$

There is another acid / base equation that is equally important and that is the dissociation of molecular water:



It provides a link between hydrogen ion and hydroxyl ion concentration in solution.

Each of the first two chemical equations detail a condition where an odorous compound can be transformed to a different compound that has no effective odour. The transformation effect relates to the bonding effect of ionic compound to aqueous environment.

The desire to drive the Henry's Law constant to zero can be achieved if the liquid phase concentration of either ammonia or hydrogen sulphide is maximised to the ionised form by the changing of pH has a similar impact to changing the characteristic of the Henry's Law constant to zero.

The relationship between pH and hydrogen ion and hydroxide ion concentration is:

$$[H^+] = 10^{-pH} \text{ and } [OH^-] = \frac{10^{-pK_w}}{[H^+]} = 10^{(pH-pK_w)}$$

It is therefore a very simple calculation that allows pH to be used to determine the relative concentration of any of the ionic species. Looking at the case of hydrogen sulphide:

$$\frac{[H^+][HS^-]}{[H_2S]} = 10^{-pKa_1} \text{ and } \frac{[H^+][S^{2-}]}{[HS^-]} = 10^{-pKa_2} \text{ also } \frac{[H^+]^2[S^{2-}]}{[H_2S]} = 10^{-pKa_2-pKa_1}$$

Looking at the total of the three compounds:

$$[HS_{Total}] = [H_2S] + [HS^-] + [S^{2-}]$$

The total sulphide concentration will be sourced from the gas phase and divided into these three compounds based on the pH of the solution. The main point being only H₂S is the only one of the three that generates odour.

By applying the equilibrium equation, it is now possible to find a ratio between soluble hydrogen sulphide and total sulphides in solution.

$$[HS_{Total}] = [H_2S] + \frac{[H_2S]10^{-pKa_1}}{[H^+]} + \frac{[H_2S]10^{-pKa_2-pKa_1}}{[H^+]^2}$$

Substituting pH relationship for the Hydrogen Ion and taking hydrogen sulphide out as the common factor

$$\frac{[H_2S]}{[HS_{Total}]} = \frac{1}{1 + 10^{pH-pKa_1} + 10^{2pH-pKa_2-pKa_1}} = \frac{1}{1 + 10^{pH-7.045} + 10^{2pH-26.045}}$$

The last factor is so small in the normal operating range, it can be neglected.

$$\frac{[H_2S]}{[HS_{Total}]} = \frac{1}{1 + 10^{pH-7.045}}$$

$$\text{As } Cg^* = H[H_2S] \text{ then } Cg^* = \frac{H}{1 + 10^{pH-7.045}} [HS_{Total}] = H_{Apparent} [HS_{Total}]$$

$$H_{Apparent} = \frac{H}{1 + 10^{pH-7.045}}$$

When pH is > 9 then the apparent value approaches zero.

This is the reason that in practice hydrogen sulphide scrubbers are designed using caustic to elevate the pH. The driving up of the pH value above 9.5 greatly improves the performance of the scrubbing system.

Similarly, for ammonia

$$\frac{[NH_4^+][OH^-]}{[NH_3]} = 10^{-pKb} \text{ and } \frac{[NH_4^+]10^{-pKw}}{[NH_3][H^+]} = 10^{-pKb} \text{ or } \frac{[NH_4^+]}{[NH_3][H^+]} = 10^{pKw-pKb}$$

Simplified further

$$\frac{[NH_4^+]}{[NH_3]} = 10^{-pH+pKw-pKb} = 10^{9.2453-pH}$$

Looking at the total of the two compounds

$$[NH_{Total}] = [NH_3] + [NH_4^+]$$

The total ammonia concentration will be sourced from the gas phase and divided into the two compounds based on the pH of the solution. Only NH₃ component of the total ammonia actually generates odour.

$$[NH_{Total}] = [NH_3] + [NH_3]10^{9.2453-pH}$$

Substituting pH relationship for the Hydrogen Ion and taking ammonia out as the common factor

$$\frac{[NH_3]}{[NH_{Total}]} = \frac{1}{1 + 10^{9.2453-pH}}$$

As $Cg^* = H[NH_3]$ then $Cg^* = \frac{H}{1 + 10^{9.2453-pH}} [NH_{Total}] = H_{Apparent} [NH_{Total}]$

$$H_{Apparent} = \frac{H}{1 + 10^{9.2453-pH}}$$

When pH is < 5 then the apparent value approaches zero.

Nearly all ammonia scrubbers are made up with either Sulphuric or Hydrochloric acid. The selection of an appropriate acid is based on economics. Phosphoric Acid has been used in the past to generate ammonium phosphate fertiliser.

7 Are caustic and acid only Scrubbers practical for design?

There are significant limitations to using only acid and only caustic chemical additions for scrubber operation. Reversibility of reaction is what makes this process impractical and the lack of affinity of the contaminant to water at neutral pH.

The examples used are a contrast into what can and what can't be achieved using simple acid / base scrubbing systems.

Looking at the Henry's Law constant for each compound:

Compound	Henry's Law Value at 20° C	Units
Hydrogen Sulphide	483	Atm/(mol/mol)
Ammonia	0.7413	Atm/(mol/mol)

It is perfectly feasible to operate a Caustic Scrubber at pH of 13.5 to remove 99% of the Hydrogen Sulphide present in a gas stream. The purge will then be returned via a purge line to the inlet works or into a sewer where the pH of the liquor as it is diluted is reduced back down towards pH 7. The end result is a re-conversion of H₂S back in solution and eventually back into the gas phase.

In the case of ammonia, the same problem exists, but only in a theoretical sense. In practice, it is much easier to maintain ammonia in solution.

When compared with Hydrogen Sulphide, Ammonia has a greater affinity to water by way of its lower Henry's law value.

Contrasting further when you consider the Apparent Henry's Law value as compared to the actual pH value at neutral pH (pH=7)

$$\text{Hydrogen Sulphide } H_{App} = \frac{483}{1 + 10^{-0.045}} = 254 \text{ atm}/\left(\frac{\text{mol}}{\text{mol}}\right)$$

$$\text{Ammonia } H_{App} = \frac{0.7413}{1 + 10^{2.2453}} = 0.00419 \text{ atm}/\left(\frac{\text{mol}}{\text{mol}}\right)$$

There may not be sufficient drive in water only scrubbers to remove ammonia from an air stream to very low levels, hence the addition of an acid. However the ammonia can be contained in the water phase provided the purge is discharged into a moderate volume of pH 7 water the overall equilibrium is maintained.

It is possible to estimate the volume of water required maintain all ammonia in solution.

Assuming Acid Scrubber is maintained at pH of 5.5 and the purge volume is at the same pH the dilution ratio required of water at pH 7 would be

$$[NH_3]_1 = [NH_3]_2 \text{ therefore } \frac{[NH_{Total}]_1}{1 + 10^{9.2453-pH_1}} = \frac{[NH_{Total}]_2}{1 + 10^{9.2453-pH_2}}$$

Through dilution although the same no. of moles of total ammonia is available, the concentration has changed.

$$[NH_{Total}]_1 V_1 = [NH_{Total}]_2 V_2 \text{ therefore } \frac{V_2}{V_1} = \frac{[NH_{Total}]_1}{[NH_{Total}]_2} = \frac{1 + 10^{9.2453-pH_1}}{1 + 10^{9.2453-pH_2}}$$

For pH₁ of 5.5 and pH₂ of 7 we have

$$\frac{V_2}{V_1} = \frac{1 + 10^{3.7453}}{1 + 10^{2.2453}} = 31.45$$

A similar calculation for Hydrogen Sulphide using caustic solution, pH₁ of 13.5 diluted down to pH₂ of 7 yields a dilution ratio of 1,500,000. Not practical!

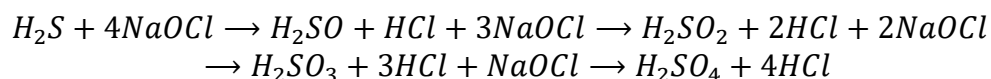
8 Addition of oxidising agents

Most common configuration of wet chemical scrubbing systems for use on Hydrogen Sulphide has been the application of Caustic with Sodium Hypochlorite onto a random packed column

The reason is that the oxidisation of Hydrogen Sulphide is a non-reversible reaction and thereby allows the by-product of the process to be discharged without risk of forming hydrogen sulphide immediately on return to neutral solution.

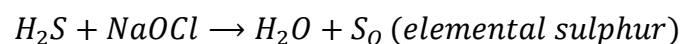
The problem is that there are a myriad of different chemical reactions and complex mechanisms at work that some understanding is required for.

There are two chemical reactions that are of key concern when designing scrubber systems:

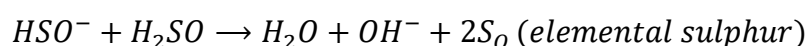
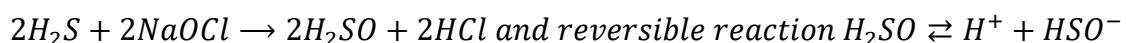


This reaction occurs at optimum pH between 9.1 and 9.3 and with ORP ranging from 620 mV to 820 mV.

The other important reaction is:



The suggested step reaction being:



This reaction occurs at pH 12.5 and ORP < 200 mV and is a problem from the point of view of practical operations as the elemental sulphur produced will very rapidly clog the packing within the scrubber column.

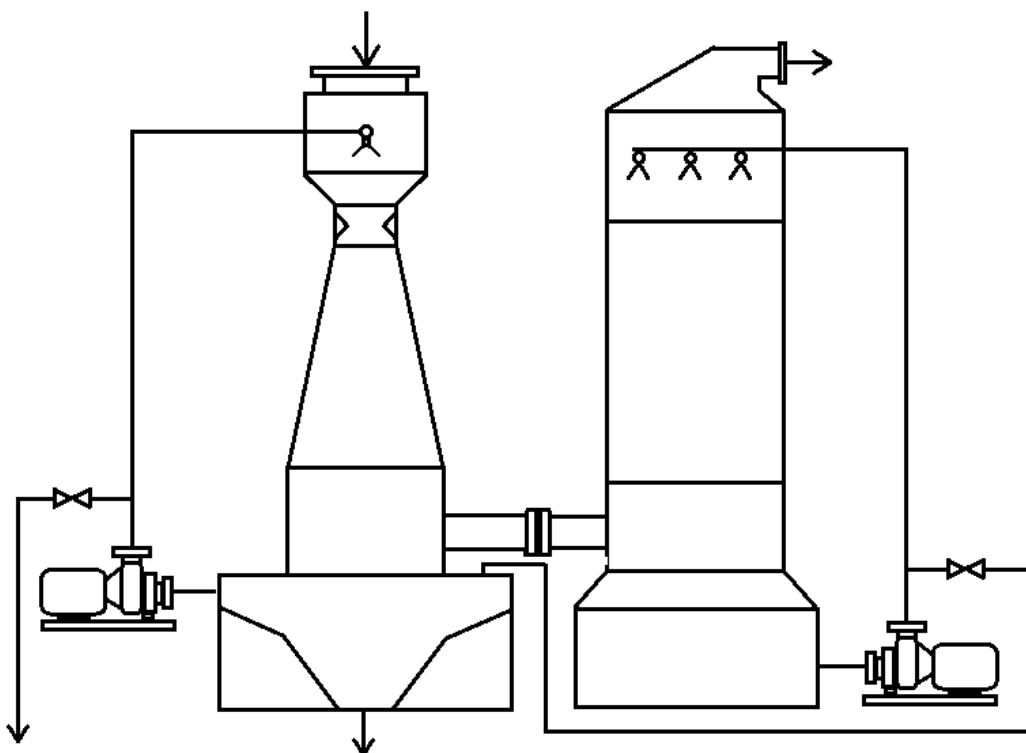
There are however some very good reasons to consider using a Venturi Scrubber, or fluidised packing scrubber that allow solids to be recovered.

Elemental Sulphur being a solid can be separated out and recovered from the waste liquor stream and sold for processing.

As noted in the chemistry, the formation of elemental sulphur uses a quarter the mass of Sodium Hypochlorite even though it uses more caustic, as caustic is cheaper the overall process is more efficient.

A combined process proposed uses a Venturi Scrubber at the front end with caustic dosing controlling the pH at 12.5 followed by a packed scrubbing column with control pH at 9.2 and ORP of 700 mV. Purge from the second tower if fed back into the first tower.

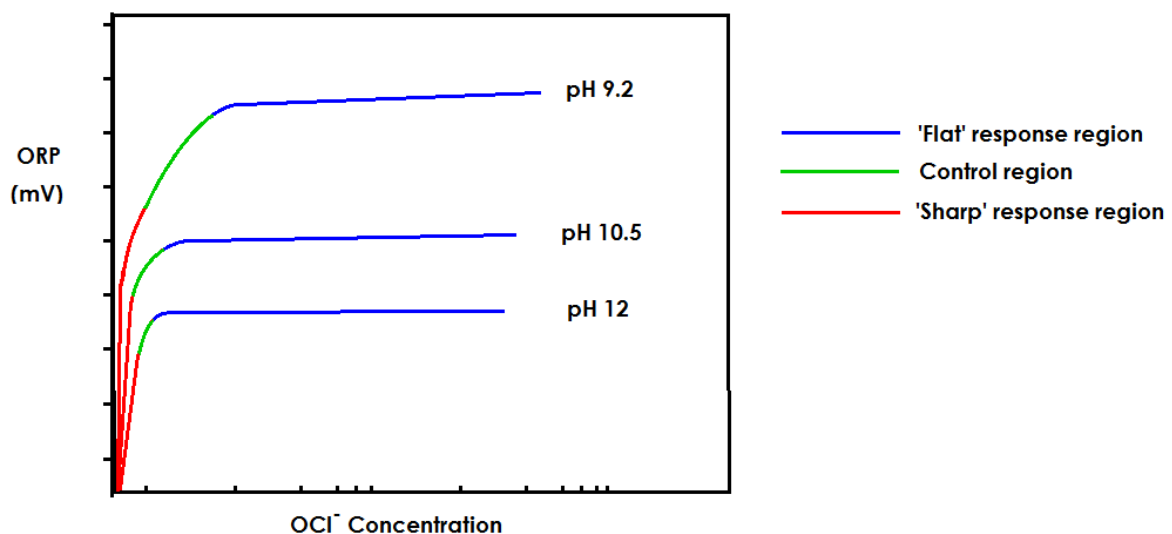
This application works well when there is a high Hydrogen Sulphide levels in the feed gas stream. The Venturi Scrubber is maintained at pH of 12.5, but has no Sodium Hypochlorite injected. The purge from the Packed Column Scrubber has some residual Sodium Hypochlorite and is sufficient to allow 80% of the Hydrogen Sulphide to convert to Elemental Sulphur.



The remainder of the hydrogen sulphide is then removed in the packed column and purged as sulphuric acid. pH and ORP are controlled in the second tower with both Caustic and Sodium Hypochlorite being injected. pH is maintained at 9.2 and the ORP to 700 mV.

The key issue to operating chemical scrubber successfully is tight control of the dosing rate of sodium hypochlorite and caustic.

If in the event caustic or sodium hypochlorite are not tightly controlled, then there is no consistent removal of hydrogen sulphide and that in extreme condition oxygen can be produced creating a hazardous situation.



pH of the scrubber liquor for the sulphuric acid formation must be strictly controlled. As can be seen from the above graph that pH of 9.2 provides a large zone where process control is achievable, whereas pH exceeding 9.2 generates a region too small for practical and reactive control.

The system can become overly responsive where in a region of sharp response there is an overshoot of ORP value with only slight addition of Hypochlorite or it enters a flat region where there is no ORP response to the addition of Hypochlorite.

The end impact is that Hydrogen Sulphide is nearly entirely converted to Sulphuric Acid in non-reversible reaction, and that the optimum column size is achieved.

9 Variables and Units

Variable	Units	Description
a, a_w	m^2/m^3	Specific Surface Areas, and Wetted Specific Surface Areas
C_P	J/kg C	Specific Mass of Fluid
ORP	mV	Oxidation Redox Potential
f	-	Fanning's friction factor
H, H_{apparent}	mol/mol	Henry's Law Coefficient, Apparent Henry's Law Coefficient (corrected for pH)
Q	m^3/s	Volumetric Flow
v	m/s	Bulk flow velocity
V	m^3	Volume
T	K	Absolute Temperature
ρ	kg/m^3	Bulk Fluid Density
[x]	mol/L	molar concentration of X in water
pKa,pKb,	-	Equilibrium coefficient based log value.
pH	-	Log Scale Hydrogen Ion concentration

10 Literature Cited in this Document

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- Technical Exchange Seminar on Odour Control Systems– MWH Australia Pty Ltd.