

CHAPTER 6:
RETROFIT AND ECONOMICS FOR NO_x ABSORPTION WITH
SELECTIVE CATALYTIC REDUCTION (SCR)

6.1 Introduction

6.1.1 An Overview

All the retrofit design options discussed in this chapter involve the scrubber/absorber. The objective is to reduce the mass load of the NO_x removal for the selective catalytic reduction (SCR). As previously explained, fewer options exist for improving the performance of the SCR, aside from purchasing newer catalysts in larger volumes. Previous chapters cover the scant improvements found for the SCR, none of which utilized equipment modification of any sort. For this reason, they do not appear in this chapter on retrofit options. The scrubber/absorber has more room for improvement and allows us much more freedom and flexibility than the catalyst vessel as far as process parameter and equipment modification is concerned.

Again, we do not claim that all of the retrofit options discussed herein are appropriate for all sites. However, the application of some of these ideas could allow the use of scrubbing at combustion sources for reducing NO. Scrubbing for NO removal has thus far proven disappointing. As a result, most sites have turned to SCR or other technologies. However, scrubbing is decidedly simpler and cheaper both in capital and operating investments due to expenses incurred in purchasing and maintaining the catalyst required for SCR. In addition, although scrubbing cannot replace SCR, it can certainly augment it. In fact, scrubbing cleans flue gases dirtied with particulates, thereby protecting the SCR from fouling. Much research remains to be done on the issue of scrubbing for NO removal and this work strives only to pique such interest.

The low flow rates of NO_x coming from the nitrocellulose plant to the NO_x abatement system may not justify the revamping options suggested here. However, the added capacity for removal by these retrofit designs may widen the application of the site from merely NO_x abatement to acid recovery. One can envision the servicing the off-gases from several other sites, including the ammonia oxidation plant and the nitric acid concentration facilities, for reducing plant-wide NO_x emissions.

The first section covers retrofit options that require little engineering design work. The first of these options involves the addition of side feeds and draws on the column. This tactic showed that we could serve the two masters of minimizing NO_x escaping the column as well as maximizing nitric acid percentage in the bottoms. The second option proposes cooling the fume stream feed to the column. Doing this, we virtually eliminate NO₂ escaping the column. However, because water condenses out of the vapor, the nitric acid concentration becomes diluted making acid recovery impractical.

The third option gives similar results to those of cooling the fume stream involves cooling the bottom acid recycle. This stream circulates within the scrubber section of the column. This only requires a chiller to cool the acid stream, whereas the second option above requires an additional heat exchanger and a secondary cooling medium (cooling water from a nearby river).

The second section covers retrofit options requiring more detailed design work. First, we explain why heat recovery is impractical for NO_x abatement at RFAAP. Next, we describe the most radical approach, that of adding a flash drum to the bottoms leaving the scrubber/absorber. This option concentrates the acid in the bottoms to recoverable levels without requiring cooling of the fumes. The third complex option is that of the addition of cooling trays or a cooling jacket to the scrubber absorber. More suited to a new site than a retrofit, this option gives similar results to cooling the fume stream at a higher capital investment.

The fourth complex option involves cooling the fume stream in two locations. The unusual NO oxidation reaction quickens at lower temperatures. Cooling to ambient conditions at the origin (the nitrocellulose line) allows more time for the reaction to proceed at a higher rate. Further cooling at the column feed promotes the exothermic equilibrium. Proposals to add ozone and/or hydrogen peroxide round out the complex design options. The addition of hydrogen peroxide to the scrubbing liquid appears to be the more promising of these two possibilities.

The conclusions section of this chapter summarizes the practical and economic considerations of the most practical retrofit options. Research proceeds vigorously for some of the ideas described in this chapter. Therefore the results presented here must remain limited for those ideas that remain to be fully explored in the literature.

6.1.2 Retrofit Considerations Particular to RFAAP

RFAAP has many spare parts (heat exchangers, tanks, piping, etc.) that we may use in retrofit designs. However, we include costs for these units in our economic calculations to maintain relevance to other sites without the benefit of such stores. Also, the use of these spares represents an opportunity cost for RFAAP.

6.2 Simple Retrofit Options

6.2.1 Side Water Feeds and Draws

Here we attempt to extend the single column to simulate the capability of two or more separate columns. This objective improves the overall performance as well as the flexibility of the existing column. The same could be done by adding additional absorption towers at extra cost, of course. For a new plant, multiple columns should be considered.

Recall from Chapter Four that the reactions driving NO_x absorption occur primarily in the top and bottom of the column. The middle section of the column exhibits little reaction. Side streams offer a solution of how to rectify this inactivity in the middle section. We show in Chapter Four that we can reduce NO_x by simply feeding more filtered water to the top of the column. However, this expedient dilutes the acid in the bottoms and also poses a column-flooding risk. By using side streams, we can optimize the acid concentration in the effluent, while still minimizing the NO_x exiting the top of the column. Figure 6.1 shows the layout and results from simulation of one possible retrofit design. Table 6.1 at the end of the chapter summarizes the total costs for this and other retrofit options considered in this chapter.

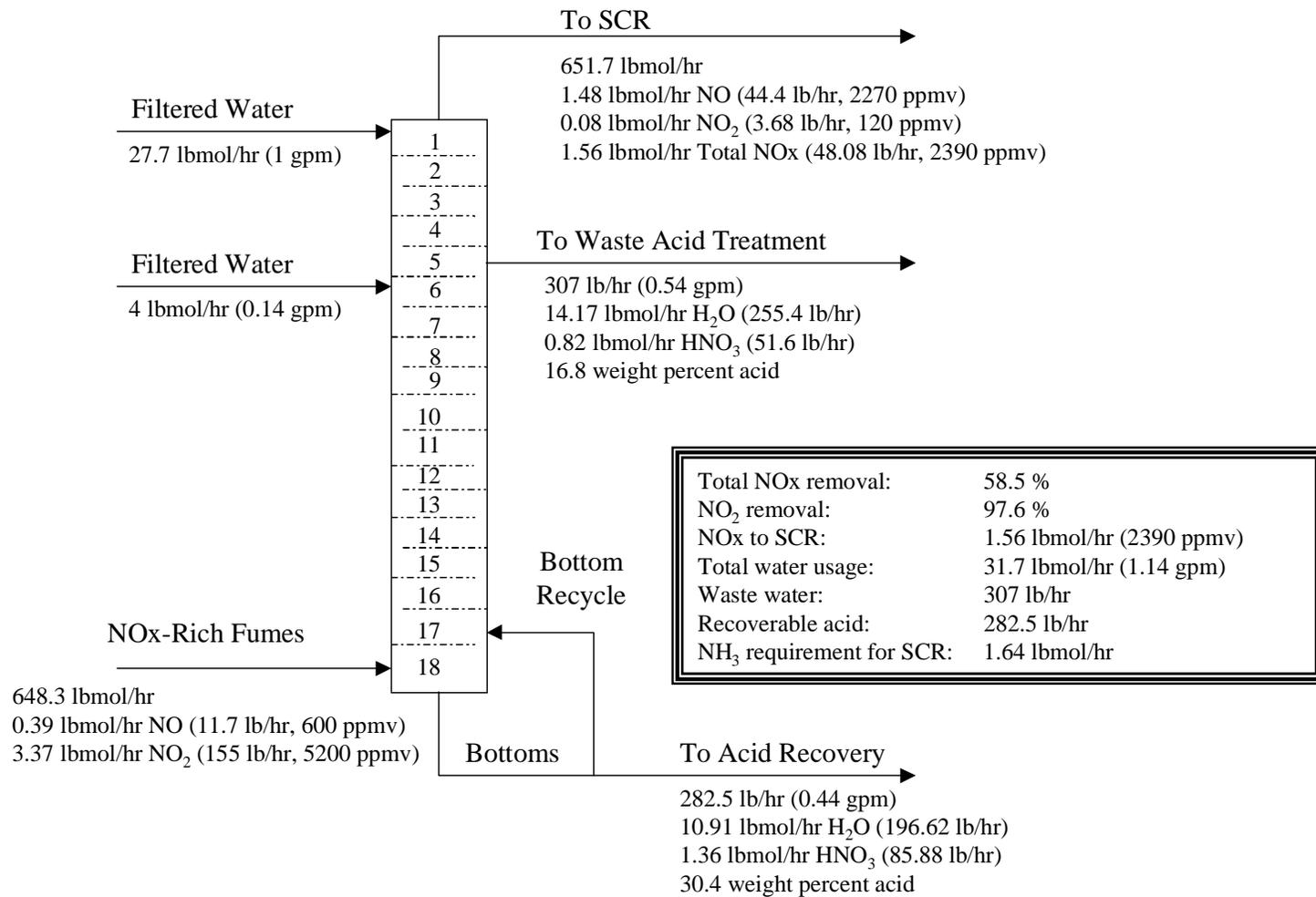


Figure 6.1. Flowsheet showing the installation of the filtered-water side streams.

6.2.2 Cooling the Fume Stream

Cooling the fume stream offers improvements in the removal of NO and NO₂. Adding a heat exchanger to the stream presents the most straightforward retrofit for this objective. Such a heat exchanger requires a dedicated chiller to cool the heat-exchange medium.

We assume the chiller yields 40 °F cooling water at 7 gallons per minute. The water to be chilled originates from a nearby river at an average temperature of approximately 70 °F. As stated above, we assume that the chiller reduces the water temperature to 40 °F. Such a chiller requires 6 tons of refrigerant. Figure 6.2 shows the block flow diagram including the chiller and heat exchanger assembly.

We choose a fumes feed temperature of 60 °F to the scrubber/absorber. We must assume attributes for heat transfer equipment. We use an approximate overall heat transfer coefficient for air to water through a steel tube wall. We approximate the individual heat transfer coefficient of the fumes with that for air. We recommend running the chilled water through the tubes at a fluid velocity of at least 3 feet per second to minimize scaling and to expedite cleaning. We also recommend using stainless steel tubes to reduce corrosion from the water. Also, the nitric acid that could condense from the fumes may necessitate the use of stainless steel for the heat exchanger shell. Figure 6.3 plots heat exchanger area versus fumes cooled temperature. From Figure 6.3, utilizing the above assumptions, we find a required heat exchanger area of 525 ft². The purchased cost for such a heat exchanger comes to approximately \$15,000 purchased cost in 1990 dollars (Peters and Timmerhaus, 1991). We use the Marshall & Swift Chemical Engineering Cost Index from *Chemical Engineering Magazine* (August 1999) to give a dollar value of \$16,428 for the second quarter of 1999. The dedicated chiller cost comes to approximately \$12,000 (1990), or \$13,142 (2nd qtr. 1999). The pump for the chilled water costs approximately \$1000 (1990), or 1095 (2nd qtr. 1999). Total piping cost including installation for

approximately 100 feet of pipe comes to approximately \$1500. Table 6.1 summarizes the total costs for this and other retrofit options considered in this chapter.

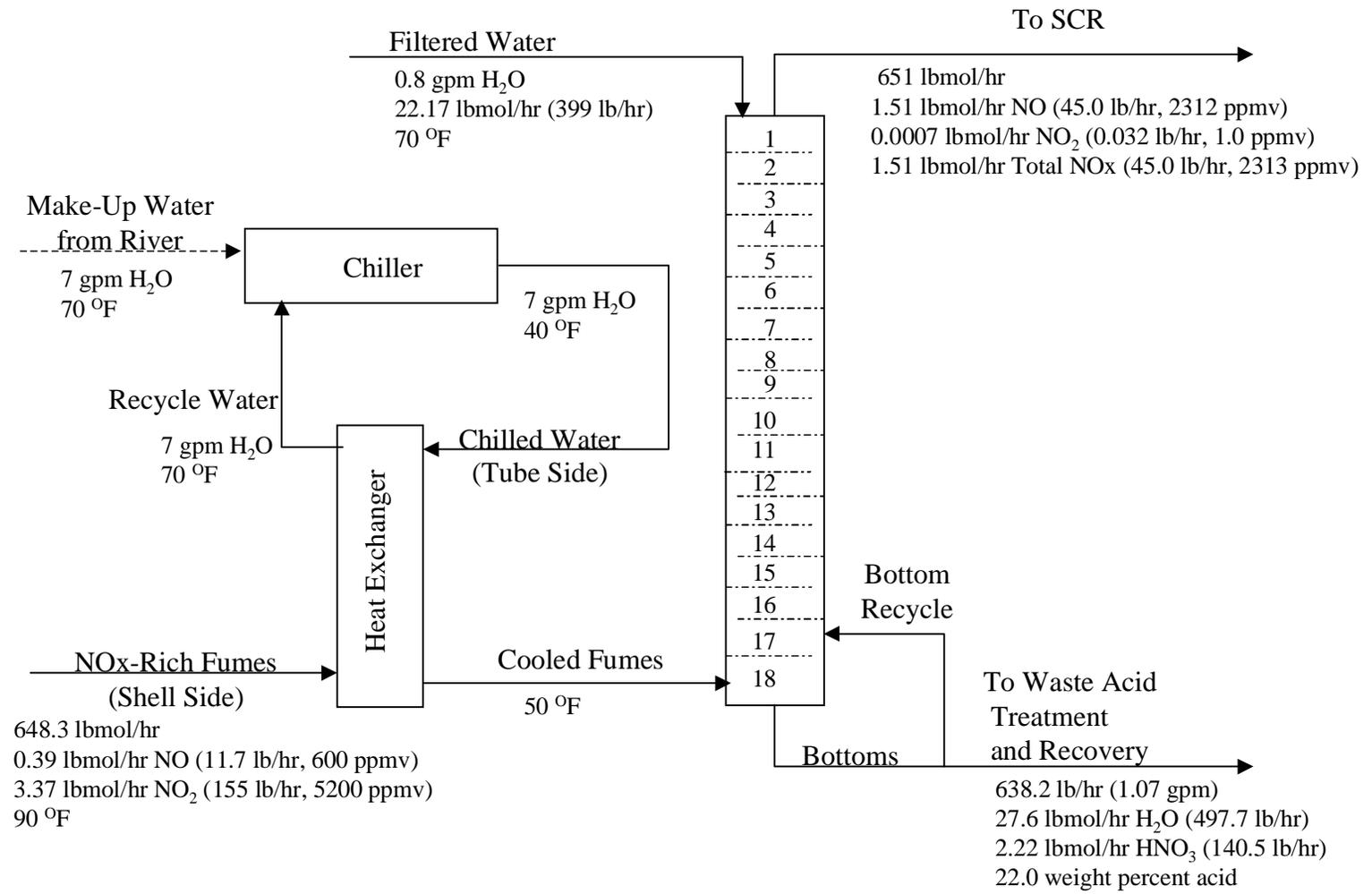


Figure 6.2. Block flow diagram of NOx abatement system utilizing fumes cooling.

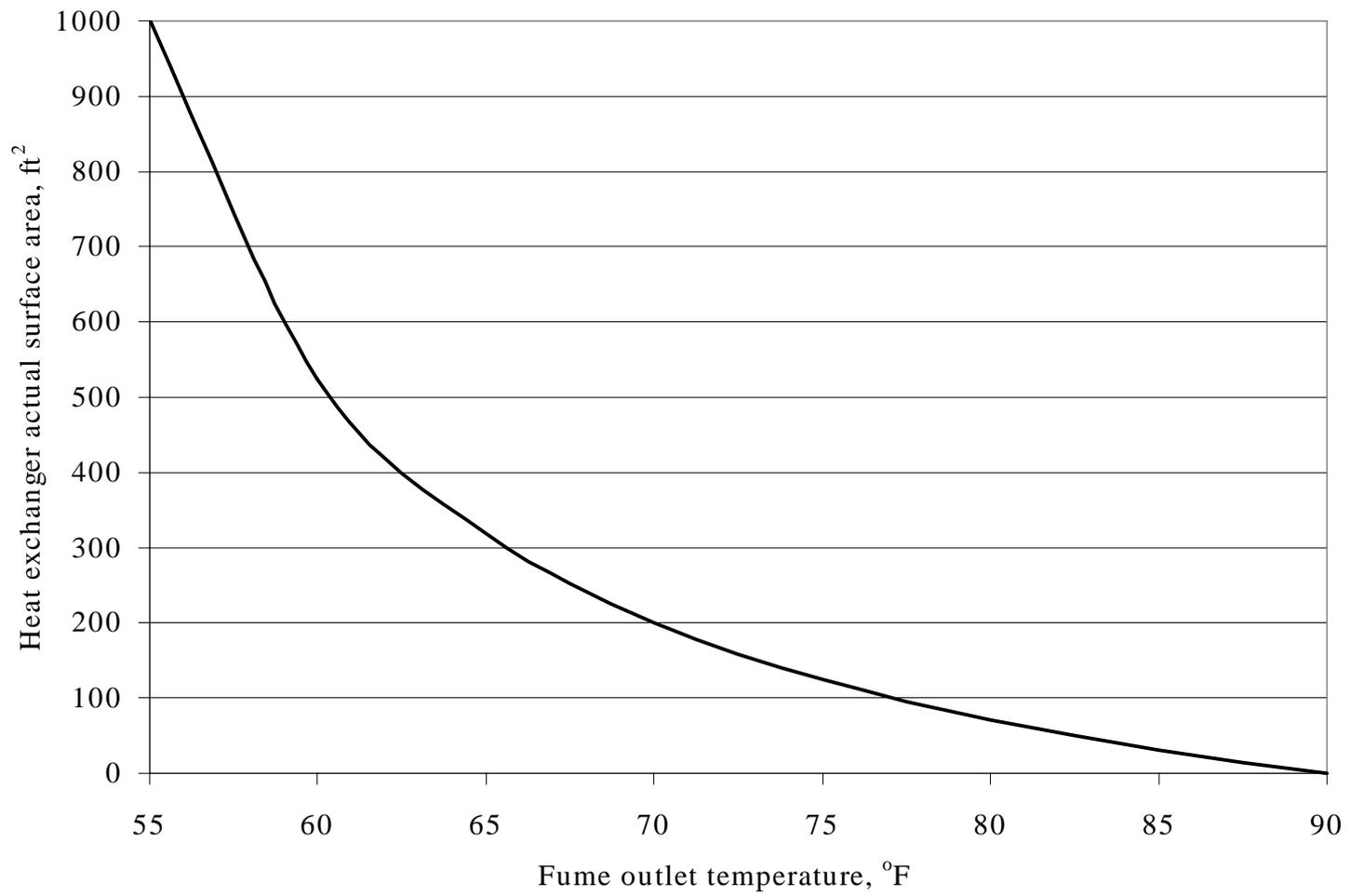


Figure 6.3. Plot of the heat exchanger area versus cooled fume temperature.

6.2.3 Cooling the Bottom-Acid Recycle

The unique construction of the scrubber/absorber at RFAAP gives an opportunity to implement a cooling strategy that may prove more efficient than cooling the fumes. The bottom-acid recycle has a high liquid flow rate, approximately 636 lbmol/hr of weakly acidic water. Chilling this flow provides a comparable molar flow rate to that of the fumes. Using this stream allows us to reach a lower overall column temperature, because we are cooling the water directly with the chiller and do not have to subsequently cool the fumes. Figure 6.4 shows the application of this retrofit design to the system at RFAAP. This option requires the presence or installation of a high-capacity scrubber like that found at RFAAP.

Installing the chiller in the acid recycle line gives several advantages. First, it eliminates the need for cooling water from the river and the additional piping it requires. Second, it eliminates the need for a heat exchanger for the fumes because the scrubber section itself acts as the heat exchanger. Third, the chilled water is sprayed directly into the fume stream of the scrubber. The spray nozzles of the scrubber supply a water mist that contacts the fumes directly with excellent heat-transfer area. Maintenance costs are reduced because such a design eliminates the need for periodic cleaning of a heat exchanger. The chiller used in this application must be capable of processing up to 40 percent by weight of nitric acid. This stipulation adds cost to the design because stainless steel must be used in its construction. Table 6.1 in section 6.4 summarizes the total costs for the retrofit options considered in this chapter.

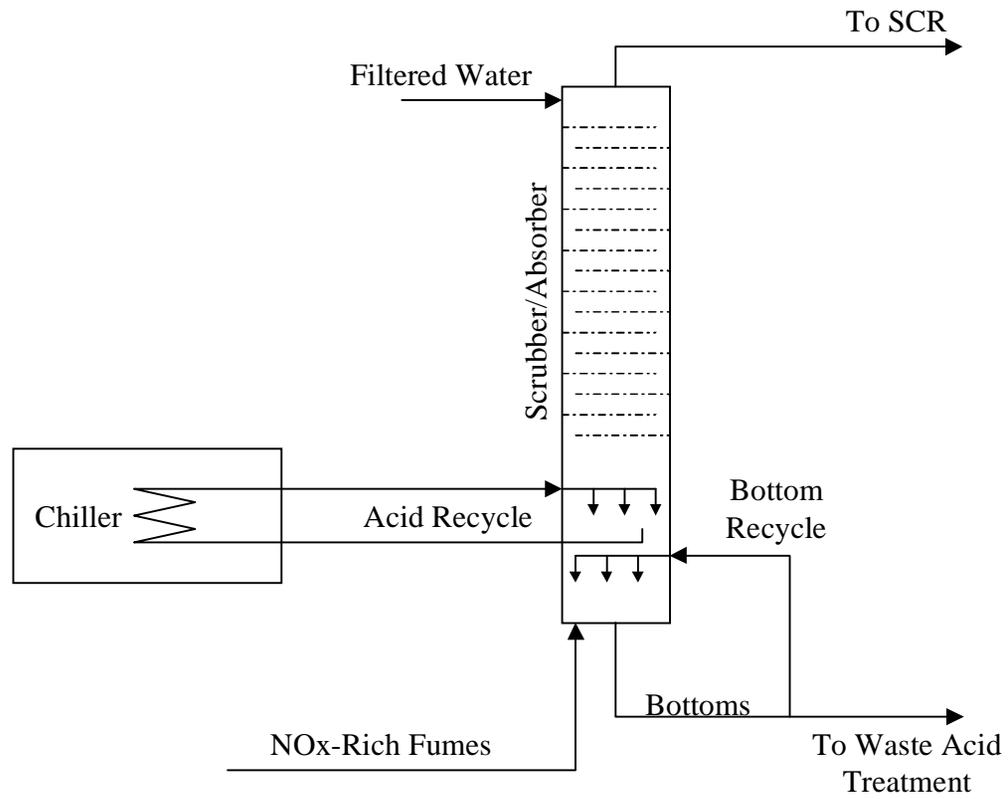


Figure 6.4. Block flow diagram of NO_x abatement system utilizing chilled acidic water recycle.

6.3 Complex Retrofit Design Options

6.3.1 Heat Recovery

The NO_x abatement process at RFAAP offers little opportunity for additional energy recovery. The integral feed/effluent heat exchanger already recovers 1.4 million BTU/hr of energy from the stack gas. After leaving the feed/effluent heat exchanger, the temperature of the stack gas is approximately 350 °F. We must maintain this temperature to minimize hazardous ammonium salt deposits in the stack. The NO_x abatement system sits 40 yards from the nearest processing plant on the RFAAP site. Therefore, piping costs for process integration with nearby processes prove prohibitive. For other sites, process integration should always be considered.

6.3.2 Acid Distillation

The acid-recovery plant at RFAAP requires a minimum of thirty percent acid by weight. The NO_x abatement system produces acid more dilute than this value and attempts at concentrating the acid in existing equipment are disappointing. The conditions of the scrubber/absorber are that NO_x mingles with very large volumes of air at very low pressure, nitric acid reacts with NO to make NO₂, and water condenses, therefore diluting the acid. Because of these conditions, thermodynamic equilibrium prevents us from exceeding thirty weight percent acid, while maintaining low NO_x gas emissions.

Previously, we addressed the difficulties of pressurizing gas streams. However, once in the liquid and isolated from NO, nitric acid can easily be pressurized through the use of centrifugal pumps. Ample heat for boiling the acid stream flows from the catalyst vessel to the stack. The heat required to boil the low acid flow rate does not decrease the stack gas temperature to the point where production of ammonium salts becomes a hazard. We merely require a simple flash drum and piping to raise the acid concentration to acceptable recovery levels. Figure 6.5 shows a design flowsheet incorporating this retrofit with key results from the

ASPEN simulation. Table 6.1 summarizes the total costs for this and other retrofit options considered in this chapter.

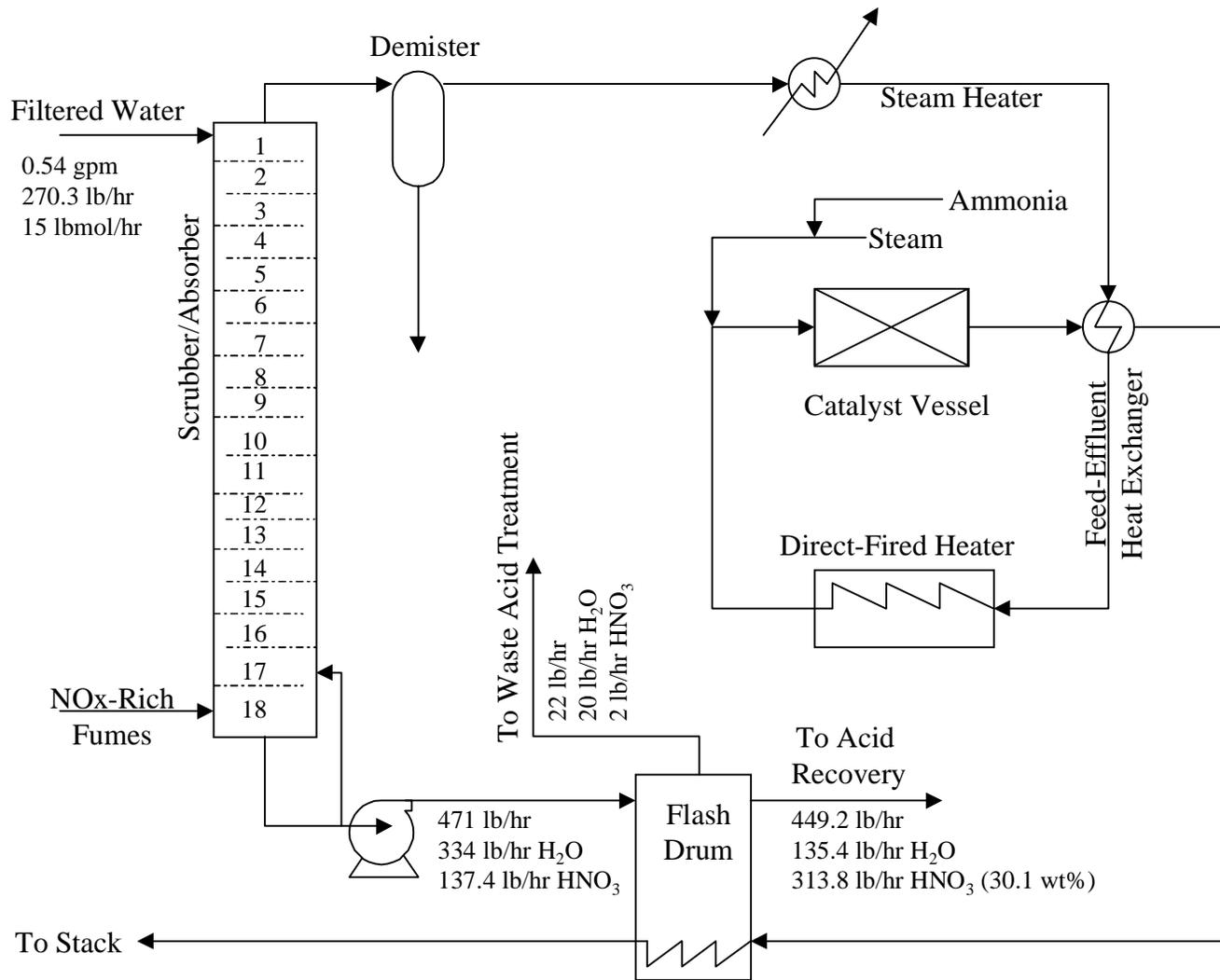


Figure 6.5. Block flow diagram of NO_x abatement system implementing acid flash.

6.3.3 Cooling Trays or a Cooling Jacket

Cooling the column directly presents an advantage over cooling the input streams, that is, we remove the heat from where it is produced by exothermic reactions in the column on the stages. Retrofitting an existing column for cooling trays or a cooling jacket represents a major undertaking. For this reason, we recommend this option primarily for new installations.

External cooling coils offer a simpler and possibly more effective option to cooling trays or a cooling jacket. Cooling trays are expensive to fabricate and more expensive to install. A cooling jacket only provides appreciable cooling to the portion of a stage where liquid touches the column wall. In the absorber tower, the tray spacing is 24 inches, and the weir height is 4 inches so the liquid level takes up a small fraction of the stage height.

The use of bubble-cap trays disallows the installation of internal cooling coils. However, we can install external cooling coils on the outside surface of the column corresponding to the level of the liquid on each stage and in the scrubber drum basin. We can purchase special heat-transfer adhesive that can help attach the coils to the column and also to improve the surface area and heat-transfer rate between the column and the coils. The use of this heat-transfer cement eliminates the need for expensive modifications to the column.

The advantage of this option over that of cooling the fumes lies in the order of magnitude larger heat-transfer coefficient for cooling water as opposed to air. Recall that we must use stainless steel heat-exchanger tubes for cooling the fumes. Also, stainless steel constitutes the column shell so the two options possess comparable wall heat-transfer coefficients. The predicted results for this option are comparable to those for cooling the fumes. We have merely eliminated the need for the heat exchanger in the fume stream in favor of the coils in contact with the column.

6.3.4 Alternative Approach to Cooling the Fume Stream

The rate of reaction of NO oxidation to NO₂ proceeds very slowly under all conditions. Temperature and pressure present the most reasonable methods by which we can increase the rate and conversion of key reactions. The duct, which carries the fumes from the nitrocellulose line to the NO_x abatement facility, represents a large, uncontrolled, plug-flow reactor. Cooling the fumes as they enter the duct maximizes conversion from NO to NO₂ in the large residence time afforded by this approximately 200-yard long duct. Cooling to approximately ambient temperatures at the source with further cooling at the entry point to the catalyst vessel minimizes efficiency losses to the environment. Also, any measures to increase the pressure of the fumes stream should occur at the nitrocellulose origination point (up to the safety limit for pressure in the ductwork) to maximize NO oxidation in the duct.

Condensed water in the duct can absorb NO₂ and form nitric acid. A collection tank installed in the line minimizes corrosion of the ductwork. A second heat exchanger located adjacent to the scrubber/absorber allows attainment of the desired temperature without allowing excessive heat absorbed in transit. Adding a second heat exchanger more than doubles the complexity and capital cost of the retrofit over the proposal for a single heat exchanger with marginally improved results. The small outputs seen at RFAAP do not warrant this endeavor. Figure 6.6 shows such a setup.

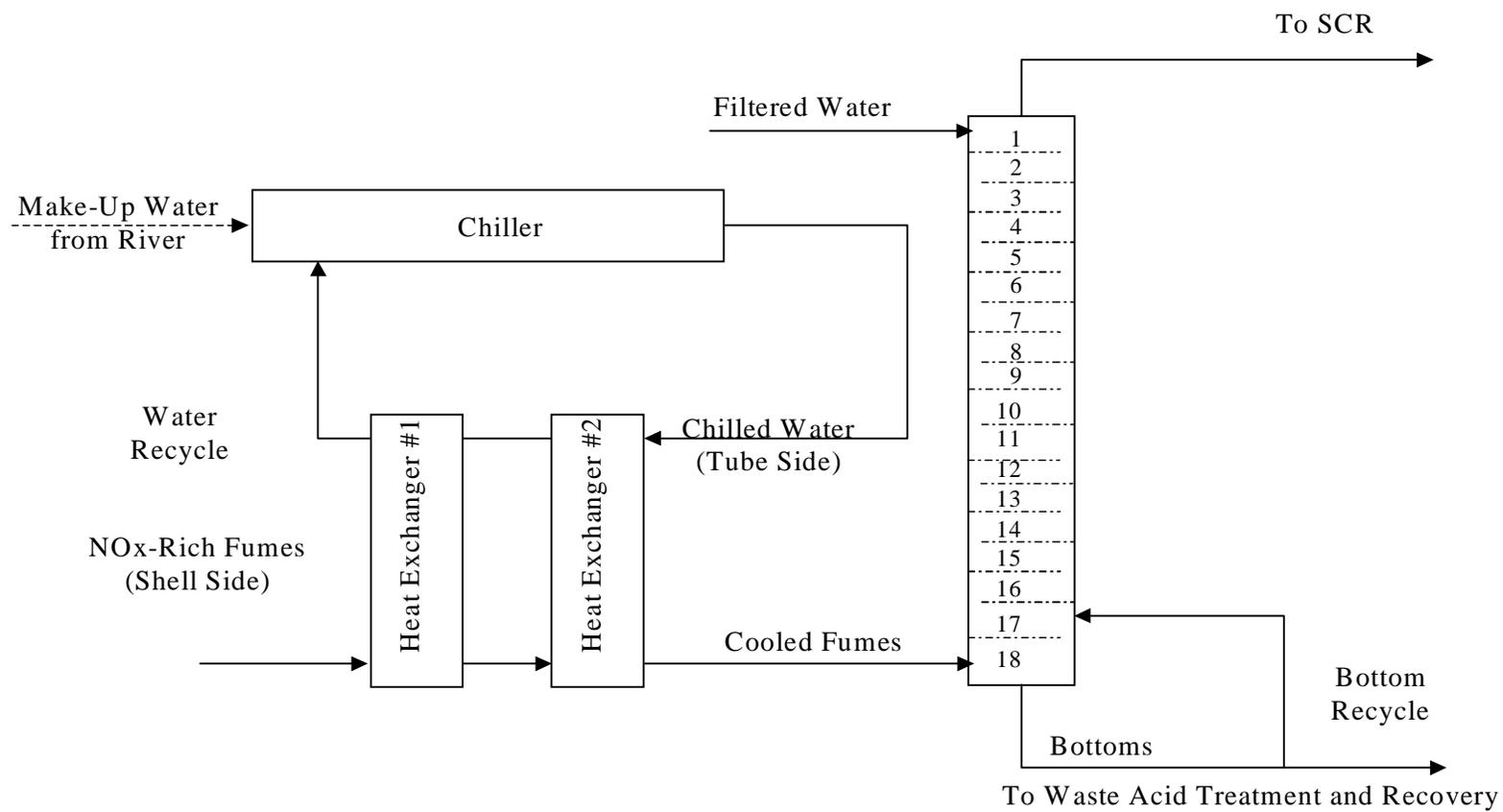


Figure 6.6. Block flow diagram of NOx abatement system utilizing dual fume-stream heat exchangers.

6.3.5 Ozone

Feeding ozone into the fumes stream rapidly oxidizes NO to NO₂ (Thomas and Vanderschuren, 1997). Use of ozone requires tight control because any release of ozone presents another hazardous emission. Also, little knowledge exists on the effect of ozone on the SCR catalyst downstream of the scrubber/absorber. Logically, ozone should improve catalyst performance because of the positive effect of oxygen.

The effect that ozone may have on the viability of the catalyst remains unknown. Until research becomes available, we should try to isolate the highly expensive catalyst from what could be too harsh of a reactant. Also, industrial ozone generators are expensive to run and operate. Therefore, we add only enough ozone to the fumes in an amount that oxidizes the initial NO feed as well as the majority of NO that NO₂ absorption produces, taking care to minimize the ozone escaping the tower. This stipulation necessitates the use of controllers to meter the addition of ozone in relation to NO_x. While presenting a potentially effective method to eliminate NO_x, ozone possesses several drawbacks. Figure 6.7 shows a hypothetical design for the use of ozone in the scrubber/absorber.

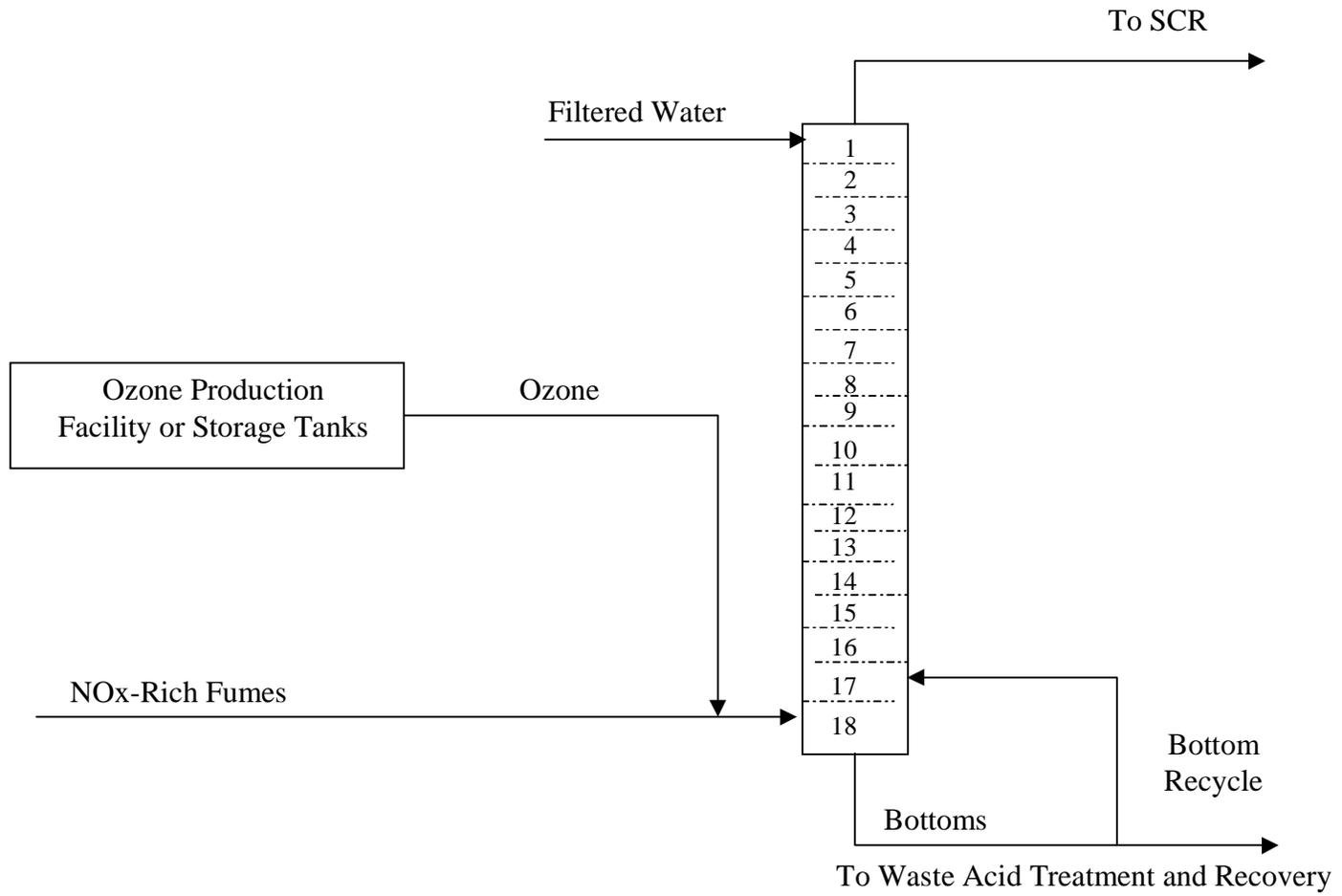


Figure 6.7. Block flow diagram of NOx abatement system utilizing ozone.

6.3.6 Hydrogen Peroxide

We discuss the effects of hydrogen peroxide in Chapters 2 and 4. We can simply feed H_2O_2 with the filtered water in a stoichiometric ratio to the NO_x fed to the column (Thomas and Vanderschuren, 1997). We can accomplish this control either through the use of NO_x analyzers or by measuring the H_2O_2 content in the bottom acid recycle to replenish that which has reacted with NO_x .

The advantage of using H_2O_2 over ozone comes from the fact that H_2O_2 remains in the scrubbing liquid. Little danger exists for its vaporizing and escaping to the catalyst vessel. Also, because it remains in the liquid, we can easily recycle it if it does not completely react with NO . Not only does H_2O_2 enhance NO removal, but it also enables us to boost the nitric acid concentration to recoverable levels.

We can combine the use of H_2O_2 and that of sidestreams to mimic a process proposed by Kuhlmann (Matasa and Tonca, 1973). The process by Kuhlmann alternates oxidation stages and absorption stages in a specially designed column. For our particular situation, we simply alternate column sections having high and low nitric acid concentrations. We add the H_2O_2 to the high nitric acid sections for oxidation of NO and the purer water expedites absorption of NO_2 . Figure 6.8 illustrates the implementation of this approach. To achieve the column shown in Figure 6.8 would require very expensive column modifications. Such an approach proves unwarranted for the low levels of NO_x seen at RFAAP.

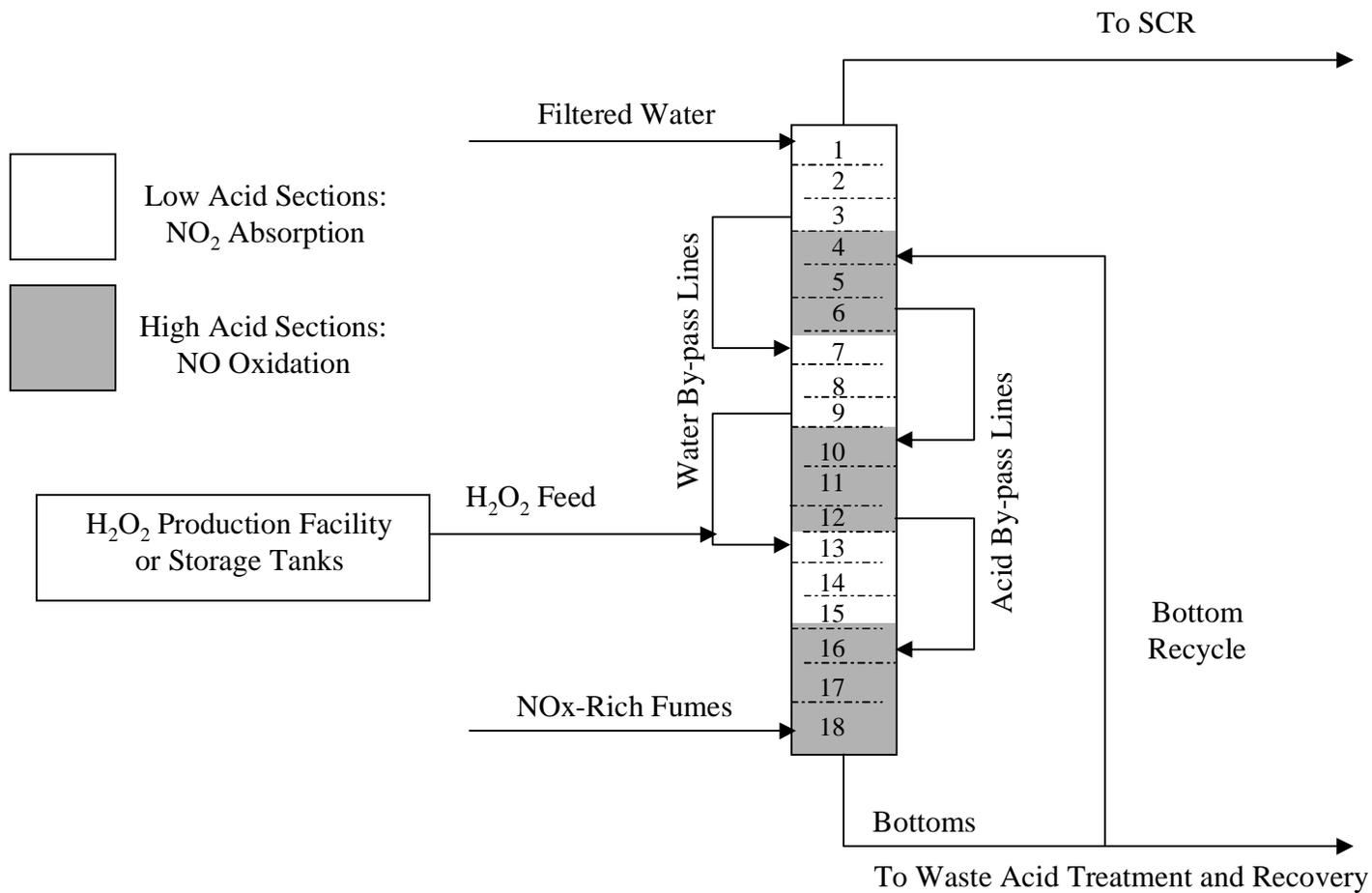


Figure 6.8. Block flow diagram of NO_x abatement system to mimic the Kuhlmann process and incorporate H₂O₂ (Matasa and Tonca, 1973).

Figure 6.9 displays the more reasonable approach to using H_2O_2 for existing absorption columns. We propose the injection of H_2O_2 near the bottom of the column where the nitric acid concentration is highest. Research shows this to be the area of greatest effect (Thomas and Vanderschuren, 1997). We require further research as to which stage constitutes the optimum injection point. We could not develop a satisfactory model for ASPEN simulation of this approach. Therefore, any results are purely speculative. However, the literature data is promising so we present some preliminary economics on the subject. Table 6.1 summarizes the total costs for the retrofit options considered in this chapter.

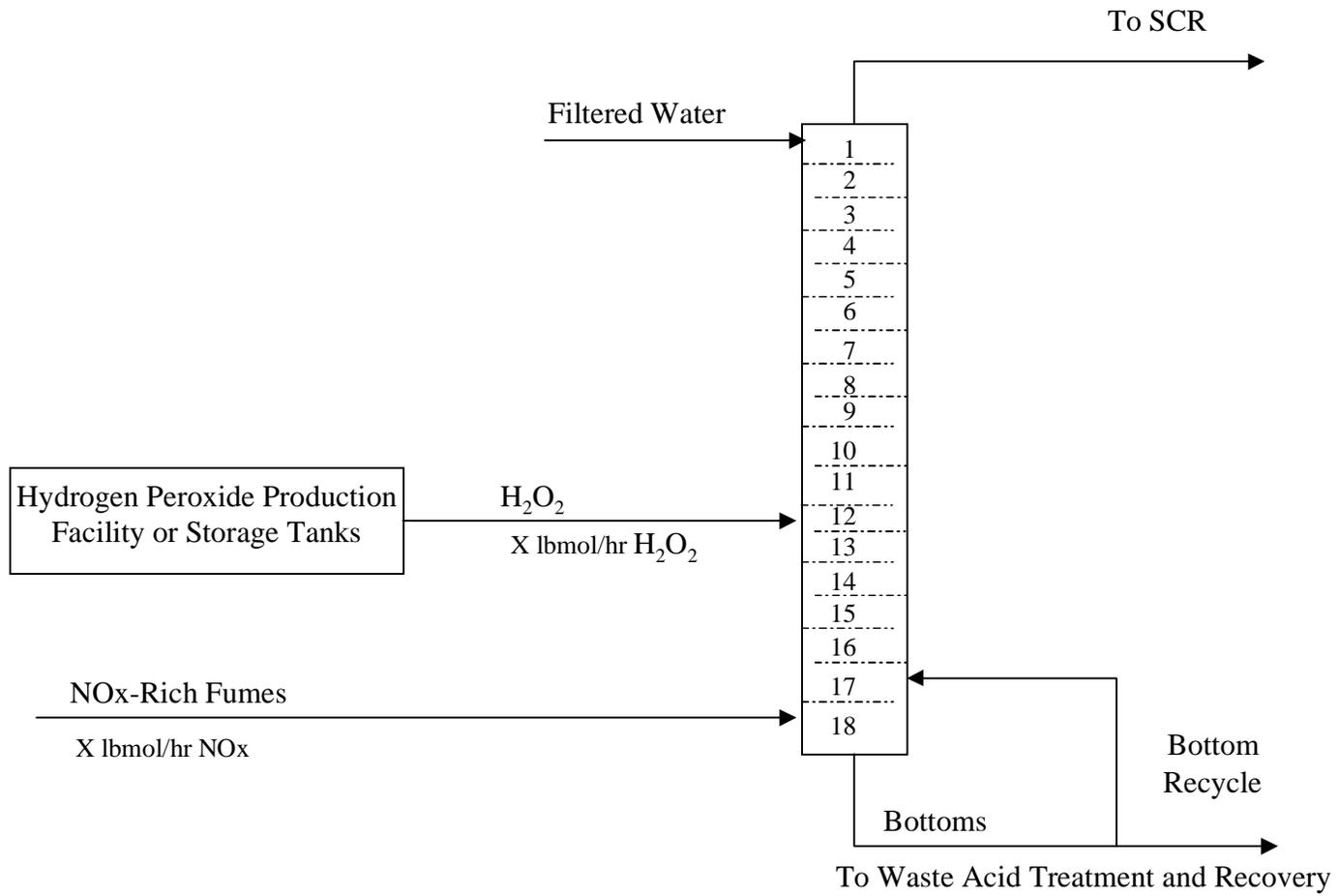


Figure 6.9. Block flow diagram of NO_x abatement system utilizing hydrogen peroxide.

6.4 Retrofit Design Economics

The various options we discuss in this chapter cannot easily be thought of in terms of traditional payback times, because the “product” in this case is not a money-maker. On the contrary, incurred penalties from EPA and DEQ can result in fines of \$25,000 per day for NO_x emissions above the guidelines. As a result, payback time could be a matter of days. In general, the options considered here represent insurance policies against such penalties.

Table 6.1 summarizes the economics of the key retrofit options we propose in this chapter. For the purposes of calculation, we assume that base case incurs one annual “incentive” from the DEQ of \$25,000. Assuming enforcement begins in earnest, such an estimate may be conservative considering the performance of the base case. All the options we propose reduce NO_x emissions sufficiently to eliminate the threat of special DEQ attentions. We consider this avoidance as an annual saving for the option. Further savings for each option appear as reductions in process water usage, slaked lime for waste acid neutralization, and the value of recoverable nitric acid. Other options require increases in operating costs as well as capital and fixed costs for the retrofits.

Capital Investment, Costs, Savings						Base Case		6.2.1: Side Water Feeds and Draws		6.2.2: Cooling the Fume Stream					
						Required	Cost, \$	Required	Cost, \$	Required	Cost, \$				
Equipment & Fixed Costs															
Pump Tank									1095		1095				
Heat Exchanger											16428				
Chiller											13142				
Column Mod.									500						
H ₂ O ₂ Storage/Plant															
Piping (installed)						15	/	1	ft	100	1500	100	1500		
Engineering						50		1	hr	40	2000	80	4000		
Skilled labor						25		1	hr	40	1000	80	2000		
Total Equipment and Fixed Costs										6095		38165			
Annual Operating Costs															
Process Water (gal)						1.1		1000	gal	3.7E+05	403	6.0E+05	656	2.1E+05	230
Slaked Lime (lb)						43		2000	lb	1.2E+06	24695	4.5E+05	9649	1.2E+06	24752
Cooling water (gal)						0.14		1000	gal					3.7E+06	524
Refrigeration (Btu)						0.04		5,000	Btu					8.8E+07	666
Electricity (kWh)						0.05		1	kWh					1.8E+05	8635
Steam (lb)						2.57		1000	lb						
H ₂ O ₂ (lb)						100		1000	lb						
EPA fines (levied)										25000		12500			
Total Annual Operating Costs										50098		22805		34807	
Recovered Acid (lb)						5		2000	lb			7.5E+05	1802	7.5E+05	1802
Total Annual Savings												1802		1802	
Compared to Base Case												51901		51901	
Payback Time (years)												0.21		2.23	

Capital Investment, Costs, Savings					6.2.3: Cooling the Scrubber-Acid Recycle		6.3.2: Acid Distillation		6.3.6: Hydrogen Peroxide Treatment	
					Required	Cost, \$	Required	Cost, \$	Required	Cost, \$
Equipment & Fixed Costs										
Pump								1095		1095
Tank										1095
Heat Exchanger								3286		
Chiller						19713				
Column Mod.										
H ₂ O ₂ Storage/Plant										2000
Piping (installed)	15		ft		100	1500	100	1500	100	1500
Engineering	50		hr		80	4000	80	4000	80	4000
Skilled labor	25		hr		80	2000	80	2000	80	2000
Total Equipment and Fixed Costs						27213		11881		11690
Annual Operating Costs										
Process Water (gal)	1.10	1000	gal		2.1E+05	230	6.0E+05	656	3.7E+05	403
Slaked Lime (lb)	43	2000	lb		1.2E+06	24752	1.2E+05	2475		
Cooling water (gal)	0.14	1000	gal				2.3E+05	33		
Refrigeration (Btu)	0.04	5,000	Btu		8.8E+07	666				
Electricity (kWh)	0.05		kWh		1.8E+05	8635				
Steam (lb)	2.57	1000	lb				1.0E+06	2685		
H ₂ O ₂ (lb)	100	1000	lb						1.1E+06	111988
EPA fines (levied)										
Total Annual Operating Costs						34283		5849		112391
Annual Savings										
Recovered Acid (lb)		2000	lb		7.5E+05	1802	1.0E+06	2499	1.9E+06	4474
Total Annual Savings						1802		2499		4474
Compared to Base Case						51901		52597		54573
Payback Time (years)						1.54		0.25		-0.20

The retrofit incorporating column side streams gives a payback time of 0.08 years. This value represents the shortest payback time of the options summarized in Table 6.1. This option reduces slaked lime requirement and allows the recovery of a portion of the acid at the expense of an increased filtered water requirement and simple column modifications and piping. This option possesses the most tenuous NO_x removal. Fluctuations in the fume feed may disrupt the delicate balance established by the careful placement of the side streams either causing out of bounds NO_x outflows or rendering the acid unrecoverable. For this reason, we include the equivalent of half of the DEQ incentive per year.

The two options involving the cooling of the fume feed to the column returned the greatest payback times. We determine the payback time for cooling the fume stream to be 0.57 years and that for cooling the acid recycle in the column to be 0.4 years. These results stem from the great capital expenditures required by these options. Also, the diluting effect of condensation within the column reduces the recovery potential of the acid and increases the slaked lime requirement increasing the operating costs. However, these options are the most reliable in terms of NO₂ elimination yielding the lowest NO₂ escape.

Distillation of the bottom acid by recovering energy from the exiting gas stream from the SCR allows recovery of highly concentrated acid and dramatically reduced slaked lime usage at the expense of moderate capital investment and minimal increase in operating cost. The eventual concentration of the acid allows the input of greater volumes of filtered water to the column, allowing the maximal absorption of NO₂.

The use of hydrogen peroxide in the scrubbing media requires further plant study due to our inability to satisfactorily model the method in ASPEN Plus. However, we recognize the potential of the option due to the fact that researchers report the ability to absorb NO as well as NO₂, greatly improving the overall NO_x removal and lessening the burden on the SCR. Also, as nitric acid increases the positive effects of H₂O₂ on NO_x absorption, we see the possibility of the recovery of highly concentrated acid. Our proposed retrofit returned a very competitive payback time. The low cost of dilute H₂O₂ in aqueous solution, the low cost of implementation of this

design, and the high volume of recoverable acid represent the keys to the economic viability of this plan.

6.5 Conclusions

1. The retrofit option incorporating the addition of column side streams possesses the lowest capital expenditure (approximately \$6,000) as well as the shortest payback time (0.08 years) of all options considered.

2. Cooling the fume stream with a heat exchanger and chiller requires the highest capital investment (approximately \$38,000) and the longest payback time (0.57 years) of all options considered.

3. Cooling the scrubber-acid recycle requires the second highest capital investment (approximately \$27,000) and the second longest payback time (0.4 years) of all options considered.

4. The two methods involving cooling offer the greatest assurance of adequate NO_x removal.

5. Acid distillation required moderate capital investment (approximately \$12,000) and the second lowest payback time (0.10 years) due to high acid recovery rates.

6. Though not fully researched, the use of hydrogen peroxide showed promise with a moderate estimated capital investment (approximately \$12,000) and a competitive payback time (0.18 years).

7. Recovery of nitric acid represents the greatest economic factor because it gives a double benefit of reducing acid costs as well as reducing the requirement of slaked lime to neutralize waste acid.

6.6 Recommendations

1. Investigate the use of side streams in all existing columns and new designs to maximize effectiveness and flexibility.
2. Incorporate column cooling in some manner if fines pose the greatest economic threat.
3. Utilize acid distillation or hydrogen peroxide if acid recovery and waste-acid neutralization represent the greatest economic factors.

6.7 Nomenclature

qtr. quarter, as in economic quarter of a fiscal year

6.8 References

Marshall and Swift Economic Indicators, *Chemical Engineering Magazine* (May, 1999).

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Peters, M. S. and K. D. Timmerhaus, *Plant Design and Economics for Chemical Engineers, Fourth Edition*, McGraw-Hill, Inc., New York (1991), pp. 661 – 677.

Thomas, D. and J. Vanderschuren, “The Absorption-Oxidation of NO_x with Hydrogen Peroxide for the Treatment of Tail Gases,” *Industrial Engineering and Chemistry Research*, **36**, 3315 (1997).