

# NICKEL'S CONTRIBUTION IN AIR POLLUTION ABATEMENT

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# Nickel's Contribution In Air Pollution Abatement

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## ABSTRACT

The Clean Air Act (United States) of 1977 not only requires tight control of emissions of sulphur, halogens and molten salts, but also requires that all utilities, process industries and municipalities have the best available technology.

A better understanding of the process chemistry and of the performance limitations of certain materials of construction has translated into more effective design and improved operation. High-nickel alloys and specialty stainless steels are used widely in incinerators and wet scrubbers.

The adoption of incinerators requires that chrome-nickel alloys overcome high temperature oxidation, sulphidation and chlorination, while the adoption of wet scrubbers often results in severe sulphuric acid attack over a wide pH range in the presence of chlorides and fluorides. Scaling and sludges strongly accelerate the modes of attack, requiring the use of chrome-nickel-molybdenum alloys for ducts, absorbers and chimneys.

Guidelines and a number of case histories will be presented to assist the design engineer in the proper selection and use of corrosion-resisting, nickel-containing alloys.

Energy production and use are major contributors to air pollution.

Examples are the burning of sulphur-containing oils to generate heat, automobile exhaust gases, fossil fuel power plants, to cite a few. Then, of course, the disposal of municipal and industrial waste — what one cannot dispose of in rivers, sea or underground is usually burned.

Figure 1 shows an estimate of the U.S. air pollutant emission over the period 1940-1983. It is obviously necessary to progress toward energy and environmental protection goals simultaneously.

The goal is to protect the environment while maintaining economic efficiency and growth. Since the 1973-74 oil embargo, coal's share of the total energy consumption in the United States has expanded from 17% to 23%, and coal-fired power plants now generate more than half of the electricity. Power plants account for roughly 85% of domestic coal demand.

The Clean Air Act (U.S.) of 1977 has helped in dropping sulphur emissions by more than 25%. There are major efforts underway to develop new materials, instrumentation, and technologies that can improve the capture of potential coal-based pollutants, including those suspected of contributing to *acid rain*. Within this program

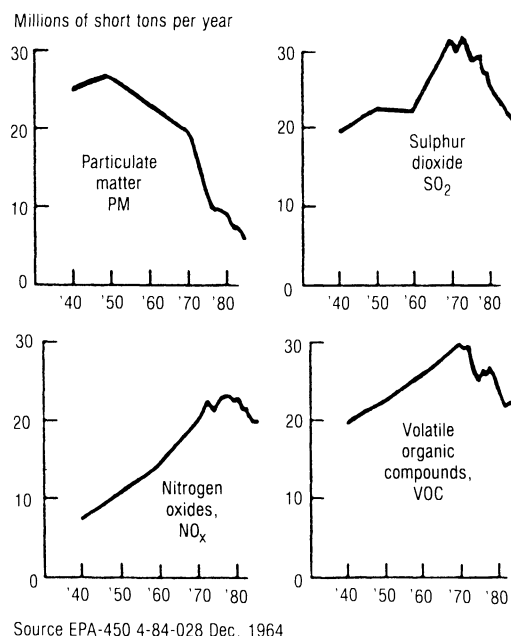


Figure 1: National air pollution emission estimates, 1940-1983

one is looking at removal of pyritic and organic sulphur from coal, and cleaning combustion gas after it leaves the boiler or gasifier.

Federal regulatory programs have imposed a cost of \$820-billion on the U.S. economy over the past ten years. As we will see, much of the equipment could not have been built and safely operated without the use of nickel-base alloys and specialty stainless steels to provide the desired corrosion resistance.

Emphasis in this article will be given to flue gas desulphurization (FGD), its technology, the corrosion problems, and the role of corrosion-resistant materials. Further, how alloys can be justified for stack liners and breeching. Waste disposal through incineration is rapidly growing, and frequently such units are linked with waste-heat recovery systems. Invariably plastics or halogens are involved, making the use of stainless steels impractical.

## FLUE GAS DESULPHURIZATION TECHNOLOGY

Significant development of FGD technology began in the late 1950s with processes ranging from the early calcium-based wet scrubbing systems to sodium carbonate, magnesium oxide, double alkali and dry collection processes. Over 75% of the plants selected a lime/limestone slurry process. There are a number of scrubbing processes utilizing a slurry of lime or limestone in water to react with the SO<sub>2</sub> and in which a sludge of calcium sulphite and calcium sulphate is removed. Process economics appear to favor limestone over lime systems.

The three main process components are the scrubber (quench section and absorber), the slurry recycle tank, and the thickener. A particulate removal section precedes the spray tower.

The hot and dry incoming flue gases containing sulphur dioxide are passed through precipitators where the solid particles, including fly ash, are removed. The flue gas at this point has a temperature of 150° C to 180° C and is essentially noncorrosive; therefore, the equipment, including fans and dampers following the precipitators, can be made of carbon steel.

The flue gas is now quenched with a water, an alkaline liquor of sodium hydroxide or a partly exhausted slurry of calcium carbonate and sulphite to form a supersaturated calcium sulphite slurry and to cool the solution down to 50° C to 65° C. The pH is brought from 1.5 to about 4 (except when using a water quench), thereby limiting the corrosivity of the medium. The presence of some oxygen converts part of the calcium sulphite to calcium sulphate. The fluid is collected in a recycle tank.

The absorber generally has several levels where the SO<sub>2</sub>-containing gas is sprayed with a lime or limestone scrubbing liquor to raise the pH to 6 or 6.5 and to absorb the SO<sub>2</sub> from the gas to form calcium sulphite. A number of demister pads in the top of the absorber eliminate droplet carryover. Generally a clear washing fluid is applied to these pads.

The slurry from the absorber is usually processed in separate storage or slurry recirculating tanks in which fresh reagents, lime or limestone, are present. After adequate residence time, the calcium sulphite and salts are withdrawn from the bottom and sent to a thickener where they are dewatered. The clarified liquor is returned to the slurry tank.

The clean flue-gas stream from the top of the absorber is now mixed with some bypass flue gas or reheated directly. A booster fan may be used to assist the discharge of the gases through the stack.

## CORROSION PROBLEMS INSIDE SCRUBBERS

The acid/chloride environment that develops inside an FGD scrubber can be extremely aggressive. The acid comes from the absorption of the SO<sub>2</sub> and small amounts of SO<sub>3</sub>. The chloride is introduced by the coal or the water in the lime or limestone absorption slurry. This environment tends to cause crevice corrosion, pitting, abrasion and erosion-corrosion. Intergranular attack of sensitized stainless steels and chloride stress cracking of AISI Types 316 and 317 stainless steels have also been observed.

All of these problems can be minimized by the effective selection and use of construction materials, by close control of the chloride level, temperature, and, most important, the pH, and by allowing a certain tolerance to deviations from standard operating conditions and design. An attempt will be made to touch on the major process factors of concern.

### Sulphur Content

The SO<sub>2</sub> and SO<sub>3</sub> form acids with water making the condensate highly corrosive. Widely varying loads (cycling and peak) and coal types (low sulphur western, high sulphur eastern and blend) have demanded variations in reagent feed rate and operating flexibility. Ideally the FGD system must be able to operate over a sulphur content range from, say, 2% to 4%, guaranteeing 90% SO<sub>2</sub> removal (95% desired).

### Chlorides

Because coals contain chlorides, their exhaust gases contain hydrogen chloride, making the otherwise protective layer of FeS porous and increasing the rate of corrosion on carbon steel.

Where the temperature of the exhaust gas falls below the dew point, moisture condenses on the metal surfaces and the acids formed with water accelerate the wet SO<sub>2</sub> corrosion. Chlorides are also present in the scrubbing water and make-up water, and concentrate due to evaporation and slurry recirculation.

In a closed single-loop system where all liquid waste streams are returned to a single reaction tank, the chloride level can build up to levels as high as 50 000 ppm, making the resultant acid chloride solutions extremely corrosive. In a two-loop system the high chloride environment can be limited to only the quencher area of the scrubber. The rest of the scrubber does not need chloride protection by high-alloy materials.

### Fluorides

Concentrations of fluorites up to 3 000 ppm have been found in the coal. They also build up due to recirculation. In particular, fluorides tend to collect under scale deposits, aggravating the acid chloride crevice corrosion on the stainless steels.

## pH Control

Close monitoring of the pH is required. To prevent sulphate scaling in the scrubber a slightly acid condition is preferred. If the pH is too low, corrosion is severe and scrubber efficiency is reduced. Optimum levels are 4.5 to 6.5 for limestone, but in the case of lime the pH can be as high as 8.0 to 8.5.

## Temperature

The amount of water in the slurry in the quench section controls the cooling of the flue gas from approximately 175° C to 50° C. One should avoid quenching the flue gas before it reaches the neutralizing slurry, and avoid the use of a freshwater quench without neutralizing agent as a pH condition of 1.5 is corrosive. In a number of cases, problems have resulted from inadequate water flow due to plugging of the water sprays raising temperatures to 90° C.

## MATERIALS OF CONSTRUCTION

In the first generation scrubbers, because of the lack of reliable alloy performance data, many design engineers had an inclination to specify carbon steel components with various coatings and rubber linings. Some of the difficulties encountered were flaking, poor adhesion, mechanical damage, erosion, and excessive temperature excursions.

First-cost savings were soon offset by maintenance and downtime. The repair of coatings and replacement linings are exceedingly more difficult operations than the initial application. Also, coatings and linings are a deterrent to making structural changes to the carbon steel vessel when welding is required.

Studies provided a good insight into the problems encountered, which range from severe pitting and crevice corrosion to intergranular corrosion and chloride stress corrosion cracking (CSCC).

Areas with excessive scale build-up, alternating wet and dry zones, high chlorides, high temperatures and low pH

are the chief zones for severe corrosion to occur, and are likely to require special stainless steels and high-nickel alloys. An overview of materials of construction capable of meeting most conditions that may be encountered in FGD scrubbers is provided in *Table 1*.

## ROLE OF STAINLESS STEELS

One should utilize the standard stainless steels where applicable, try to understand and define their limits of usefulness, and select materials containing higher molybdenum, chromium and nickel content for those areas where more severe conditions are encountered.

With today's melting practices, low-carbon stainless steels or, alternatively, alloys stabilized with titanium or columbium to provide the desired resistance to intergranular corrosion, are readily available. The problem of CSCC can be readily met by adjustment of the nickel content in the alloy.

In practice, alloys with a minimum nickel content of 22% seldom experience CSCC, while alloys with minimum nickel contents of 42% are not expected to be susceptible to CSCC. In FGD scrubbers examined so far, it appears that CSCC of the austenitic stainless steels is not a problem at the usual operating temperature of 50-65° C but may become a problem at higher temperatures. A number of failures have occurred in reheater tubes; substitution with alloy 825 appeared to solve the problem.

One would be greatly concerned on hot surfaces (wet/dry conditions) where scaling and chloride enrichment would occur. At these locations, however, the problem of excessive pitting and crevice corrosion appears to overshadow CSCC and makes the selection of the stainless steels undesirable.

The problem of pitting and crevice corrosion can be extremely severe on stainless steels in scrubber service.

The pitting index of an alloy is equal to the sum of its chromium content plus 3.3 times the molybdenum content. This formula has proved to give good correlation with actual experience in seawater systems, showing alloys with an index number of 32 to be resistant to pitting and

TABLE I

A SELECTION OF STAINLESS STEELS AND HIGH-NICKEL ALLOYS FOR CONSIDERATION IN FLUE GAS DESULPHURIZER SCRUBBING SYSTEMS													
UNS Number	Stainless steel alloys							High nickel alloys					
	316L	317LM	2205	2328	904	904hMo	28	20	825	825hMo	G-3	C-276	625
	531603	531725	531803	—	N08904	N08925	N08028	N08020	N08825	—	N06985	N10276	N36625
Nominal analysis, %													
C max	0.03	0.03	0.03	0.04	0.02	0.02	0.2	0.03	0.025	0.025	0.015	0.015	0.10
Cr	17.5	18.0	22.0	23.0	20.5	20.5	27.0	20.0	21.0	21.0	22.0	16.0	21.5
Ni	13.5	13.0	5.0	27.0	25.0	25.0	31.0	37.5	42.0	42.0	48.0	57.0	61.0
Mo	2.75	4.5	3.0	2.75	4.7	6.0	3.5	2.5	3.0	6.0	7.0	16.0	9.0
Cu	—	—	—	3.0	1.5	1.0	1.0	3.5	2.5	2.5	2.0	—	—
N	—	0.14	0.15	—	—	0.12	—	—	—	—	—	—	—
Cb/Ta	—	—	—	—	—	—	—	0.3	—	—	0.3	—	3.65
Others	—	—	—	Ti	—	—	—	Cb	A1.Ti	A1.Ti	Co	Ti	A1.Ti.Co
Pitting index	26	32	32	32	36	40	38	28	31	41	43	N/A	51
Estimated cost ratios													
Plate	1.0	1.4	1.4	1.8	2.0	2.6	2.6	2.8	2.8	3.6	4.0	6.8	5.9
Tube—seamless	1.0	1.2	1.2	1.4	1.6	2.2	1.8	—	2.2	3.1	3.5	5.0	4.3
—welded	0.5	0.6	0.6	—	1.3	—	—	1.8	1.8	—	2.1	3.8	3.3

**TABLE 2**  
**GUIDELINES IN ALLOY SELECTION**  
**IN FGD SCRUBBING SYSTEMS**

	Chlorides ppm	MILD		MODERATE		SEVERE	
		<100	<500	<1 000	<5 000	<10 000	<50 000
	pH Acidity						
MILD	pH 6.5	316L	317L	317LM	904L	904hMo	G
MODERATE	pH 4.5	317L	317LM	904L	904hMo	G	625
SEVERE	pH 3.0	317LM	904L	904hMo	G	625	Severe corrosion and pitting attack
	pH 1.5	904L	904hMo	G	625		

those with an index of 36 to be resistant to crevice corrosion.

Figure 2 provides a ranking of various alloys in their resistance to pitting and crevice corrosion. It is relatively accurate for alloys containing 16 to 28% chromium and 1 to 7% molybdenum. The higher the pitting index the more resistant the alloy is to pitting and crevice corrosion.

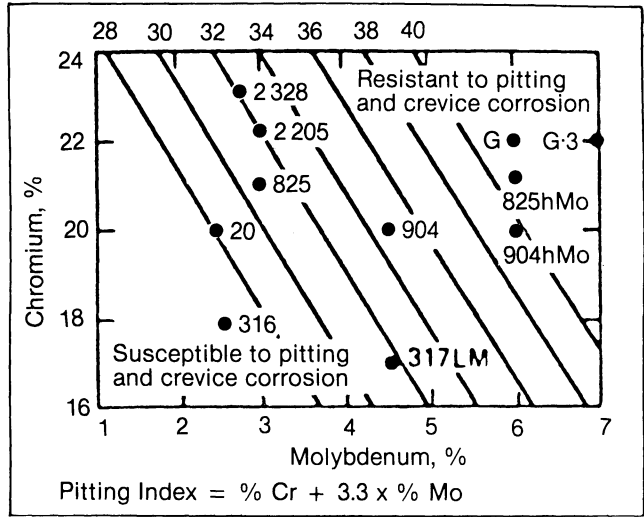
An attempt has been made to correlate the pitting and crevice corrosion obtained on Type 316 and Type 317 stainless steel test samples with chloride and pH measurements. These data have been projected and the probable performance of alloy Type 317LM plotted to reflect the difference in resistance to pitting and crevice corrosion in the temperature range of 50-65° C for the incremental 1% molybdenum content in the three alloys.

Figure 3 provides a conservative alloy selection guide for stainless steels in FGD scrubbing systems in the range of pH 3.5 to 7.0, and 10 to 10 000 ppm chlorides. Note that these stainless steels have a range of molybdenum content that can alter their exact position. For example, the Type 316L stainless usually contains 2.25% Mo, but can be specified to contain 2.75% Mo. Experience has shown that Type 304L stainless steel is inadequate in the wet portions of the scrubbers.

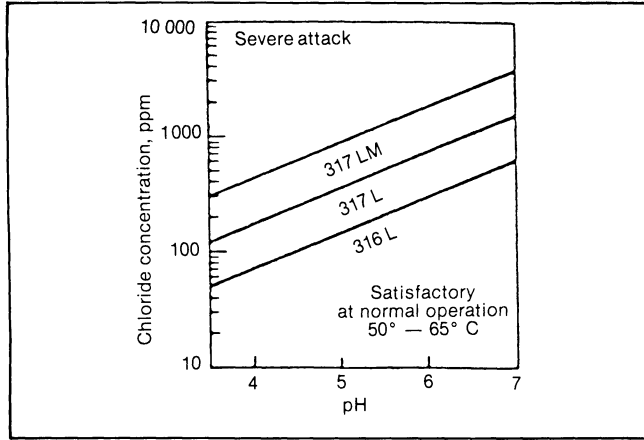
**ROLE OF HIGHER Ni-Cr-Mo ALLOYS**

From the data in Figure 3 and the various industry surveys, it can be seen that there are conditions in the scrubber under which the standard stainless steels, even Type 317LM, will not have adequate corrosion resistance. High chloride contents and low pH operating conditions are the chief causes for pitting and crevice corrosion. The beneficial effect of molybdenum in minimizing this problem has been clearly identified. A performance ranking of Cr-Ni-Mo and Ni-Cr-Mo alloys such as those in Table 1 would aid the utilities and FGD equipment suppliers in selecting materials to match a specific application.

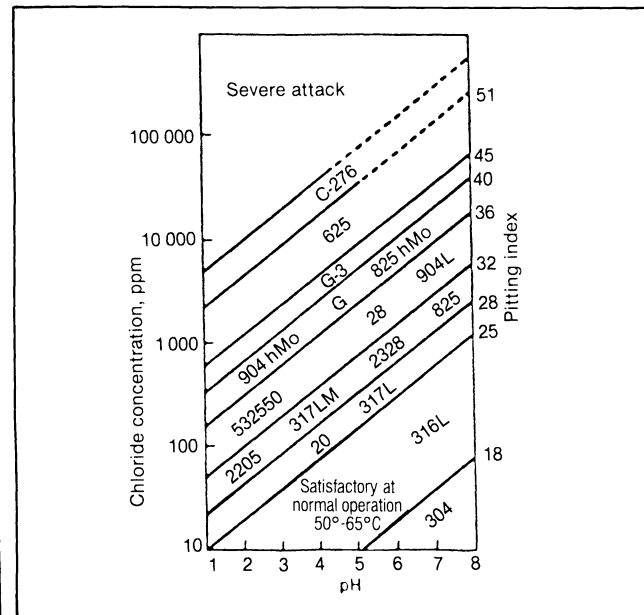
Table 2 is an attempt to define certain operating conditions as mild, moderate and severe in terms of corrosion, assuming a normal scrubber temperature of 50-65° C. This essentially establishes a range of conditions for which a certain alloy may be considered. These guidelines are plotted in Figure 4. While extensive comparison with pub-



**Figure 2:** Relative resistance to pitting and crevice corrosion



**Figure 3:** Limits of usefulness of stainless steels in FGD scrubbing systems

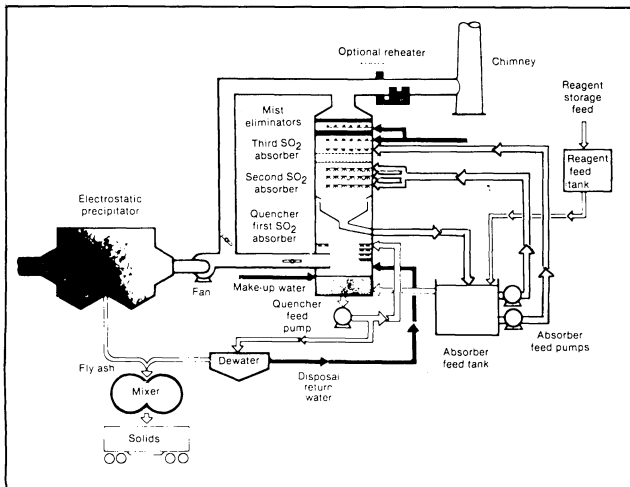


**Figure 4:** Limits of usefulness of specialty stainless steels and Ni-base alloys in FGD systems

lished information has taken place, caution is recommended as the effects of temperature deviations, presence of fluorides, unusually heavy scaling and undesirable design features or fabrication procedures cannot be interpreted in such an oversimplified graph. The environment of each flue gas scrubber is unique — a product of fuel, water and design.

### TYPICAL APPLICATIONS

There are many different designs and process control systems, therefore one can only discuss alloy selection for a specific application in general terms. *Figure 5* shows a flow diagram of the Research Cottrell system. It is shown to orient one's thinking without implication of design preference. The EPRI Report CS-1736 has been consulted in deriving the following comments.



**Figure 5:** Flow chart of Research Cottrell limestone scrubbing system for flue gas desulphurization

First, it must be recognized that a failure of the equipment downstream of the scrubber, such as the outlet duct, reheater, booster fan, breech entry and chimney line can mean a shutdown of the entire power generating station, unless one has several scrubbing units in parallel and has designed for sufficient flexibility to cope with a wide range of load.

For this reason the outlet duct is frequently constructed of alloy 904L as hot bypass gases and moist scrubber exit gases mix; for the reheater, alloy 825 or G is used to avoid CSCC. For booster fans and housing downstream from the scrubber, alloy 625 is often selected for corrosion and abrasion protection and its excellent strength and fatigue properties.

The breech entry can be made of alloy G or 625 as hot gases can aggravate corrosion here. For chimney lining alloy G or 625 offer the greatest flexibility in performance. The outlet dampers are a critical component and are frequently specified in alloy 825 and more recently in alloy 625 to resist pitting and CSCC, while the seals on these dampers are generally of alloy 625.

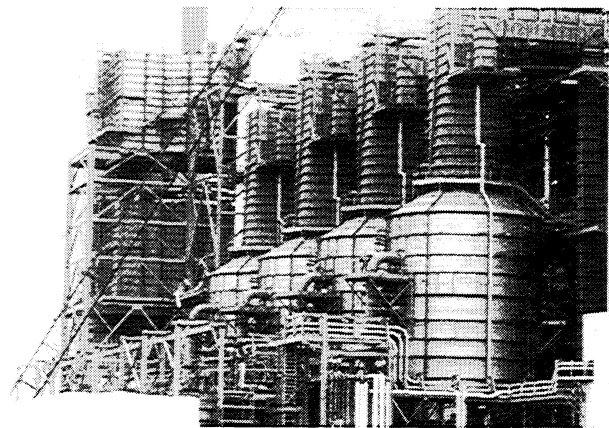
The inlet of the prescrubbers is considered one of the worst corrosion areas. This is where the hot entering gas first meets wet limestone slurry. Heavy deposits of solids high in chlorides and low in pH are most likely to form. For this reason, alloy G is generally specified here. In some cases 625 is selected for the wet/dry area to resist corrosion and abrasion. Alloy 904L is useful in the quen-

cher with alloy G for the bottom to resist chloride accumulation under deposits.

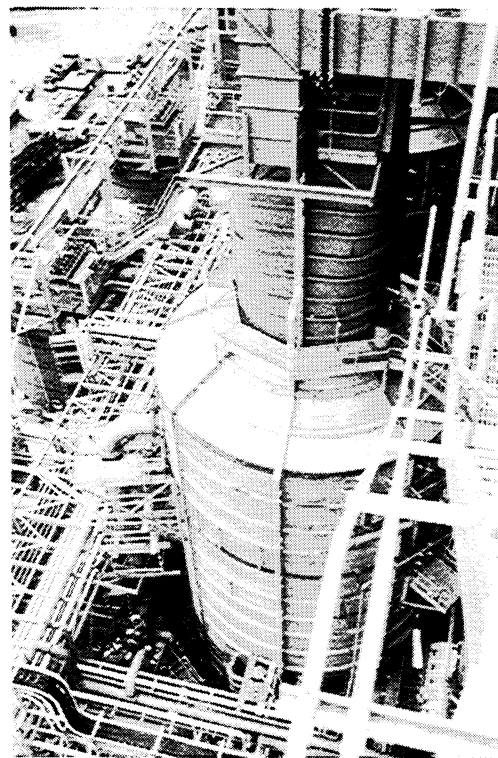
The absorber itself in a two-loop system could employ Type 316L or 317LM stainless in the upper section of the tower, for the mist eliminators and spray nozzles, and alloy 904L for middle and lower parts for the tower walls and the roofs of the nozzles.

Judicious selection of corrosion-resisting alloys can keep utility downtime to a minimum. Tampa Electric Company insisted on the best possible equipment for its new Big Bend N° 4 power station, so that the FGD reliability would match that of the boiler-turbine/generator. Corrosion tests showed how well alloy 625 withstood the highly corrosive conditions expected in the forced oxidation process. Alloy 625 was selected for the quencher section and alloy 904L in the upper half of the tower, the absorber section.

*Figures 6 and 7* show the three operating modules and

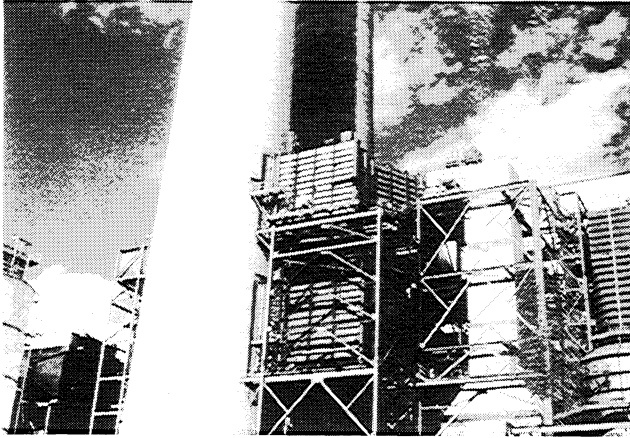


**Figure 6:** Tampa Electric Company Big Bend N°4 Station. Four quench absorber towers discharging into treated gas ducts



**Figure 7:** Individual tower — absorber of 904L; quencher, alloy 625

the one spare module included in the system. *Figure 8* shows the dual entry breach with the steam-to-clean-air heat exchanger, feeding hot air into the bypass duct to the stack.



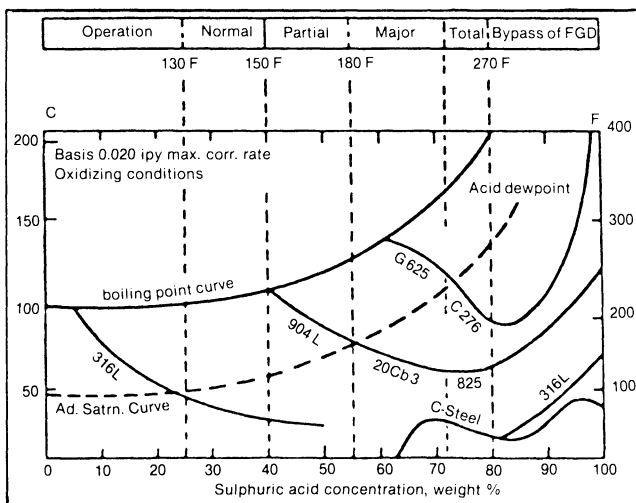
**Figure 8:** Tampa Electric Company Big Bend No. 4 Station. Dual entry breaching with steam-to-air heat exchanger

### STACK LINERS AND STACK CORROSION

The reliability and expected life of power plant stack linings is directly related to stack design, materials of construction and the flue gas atmosphere being handled. Years ago when flue gas temperatures rarely dropped below 170° C, there was little concern about acid condensation on liner walls.

The tall stack policy prevailed, putting the air pollution into the upper atmosphere and far away. With the advent of the FGD systems, gas entering the stack is a mixture of scrubbed and bypassed boiler flue gases.

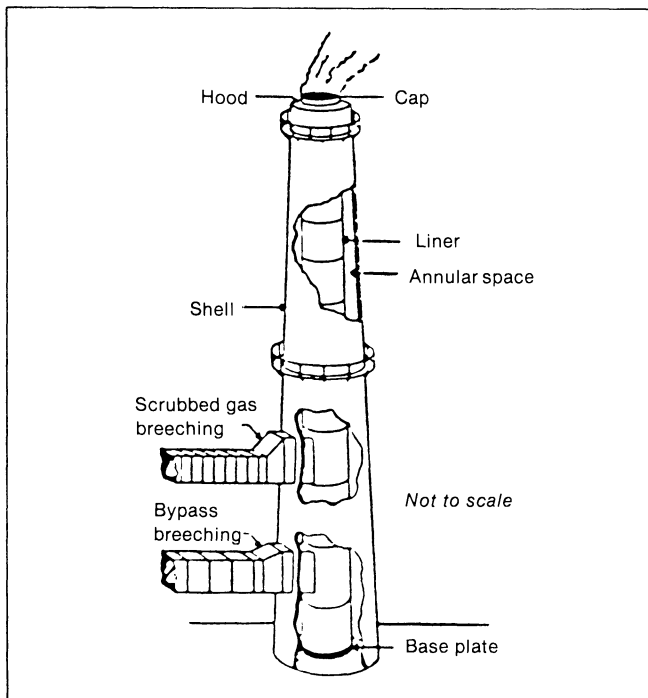
Normal operation in compliance with the emission limitation of 90% SO<sub>2</sub> removal will result in mixed gas temperatures of 55-85° C. Higher temperatures of 85-130° C may occur during start-up and abnormal conditions, when a higher percentage of sulphur dioxides in flue gas bypasses the scrubbers. These gases will normally be below the acid dew point. From the adiabatic saturation curve on sulphuric acid, *Figure 9*, one can determine the



**Figure 9:** Adiabatic saturation curve shows sulphuric acid concentration for various temperatures and operating conditions

approximate concentrations of sulphuric acid expected in the condensing droplets, ranging from 25% to 55% under normal operation and up to 80% under full bypass conditions. These are severely corrosive conditions.

*Figure 10* shows the principal components of a typical stack associated with a wet scrubber system. Stacks have either single or dual entry breeches. Breeching for bypass flue gas, being above the acid dew point of 170° C, can be made of carbon steel. Breeching from the scrubber outlet and the stack liner need to be protected from sulphuric acid corrosion. One can choose between coatings and alloys. Carbon steel coated with 60 mils of a fluoro-elastomeric or a glass-filled polyester have been used with mixed success; they have their limitations in terms of temperature, faulty application, repair, flaking and debonding,



**Figure 10:** Principal components of a typical stack

downtime and revenue loss. Stacks are often lined with acid-resistant brick. Cracks in the mortar frequently occur. To prevent acid from penetrating the cracks, the annular space should be pressurized.

For maintenance-free breeching and liners, alloy G, alloy 625 or alloy C-276 are suggested. There are a number of successful applications with alloys 625 and C-276 as stack liners. As the adiabatic saturation curve shows, temperatures in the range 115-170° C need to be avoided, and the gas temperature raised above the dew point by utilizing in-line reheat.

Alloy 625 is the most suitable selection for expansion joints. Chimney corrosion can be a severe problem when the sulphur content in the coal exceeds 1%.

### INCINERATORS

Disposal of solid, liquid and gaseous waste products through incineration has become an important and environmentally necessary practice. It is usually done by liquid injection, fume incineration, or in fixed hearth or rotary kiln equipment. Ocean incineration has been adopted for hazardous waste and sludge.



Because of burning of chlorinated compounds and plastics in chemical as well as municipal waste, the flue gases in the waste heat recovery boiler can contain several per cent HCl and lower concentrations of Cl<sub>2</sub> and salts. Sulphur and phosphorous compounds are also frequently present. The economics of waste incineration are heavily influenced by the ability to recover waste heat, usually in the form of steam. While the burning kiln can be lined with acid-resistant brick, there are many associated components such as spray towers, mist eliminators, cyclones, combustor grid supports, fans and fan housings, ductwork and dampers that require structural strength and good resistance to corrosive gases and fluid at high or low temperatures.

The stainless steels such as Types 304 and 316 play a limited role in these systems, having generally good corrosion resistance above the dew point and up to 315° C. During shutdown, stainless steel is susceptible to pitting, crevice corrosion and chloride stress cracking. Alloy 825 and alloy 600 have given superior performance and have been used up to 540° C, resisting oxidation, sulphidation and chlorination. Attack on metals underneath salt deposits may require the Ni-Cr-Mo alloys such as 625 and C-276. As each situation is different it is not possible to provide general rules.

In high temperature systems from 800 to 1 300° C, corrosion rates on waste heat recuperators can be very severe and many times the process system has to be designed so that metal wall temperatures are substantially lower to resist halogens and molten salts. As waste burning processes are rapidly expanding, more attention is being given to determining compositions and effects of emissions, resulting from incineration, and a growing role for specialty stainless steels and high-nickel alloys is visualized.

### OTHER POLLUTION CONTROL DEVICES

While wet scrubbers and incinerators offer the largest nickel potential in the power industry and for industrial waste disposal, in excess of 10 000 tons of alloy per year, numerous other applications where nickel plays a role come to mind. For example:

In electrostatic precipitators, for plates and wires alloys 800, 825, 600, 601 and 625 have been used.

In thermal and catalytic converters for control of automotive exhaust gases, in sheet and strip form, alloy 601.

In metallurgical smelters and for the the handling of waste gas systems of steel mill furnaces, alloys 825, G and 625.

And various alloys are employed in the production of sulphur and sulphuric acid from tail gas cleaning operations.

Energy technology will control much of our activities in the coming decades. In new areas of encounter, laboratory and pilot plant test programs will have to be undertaken to investigate the corrosion of alloys and the factors affecting it, such as temperature, gas or liquid state, configuration of the structure, crevices and under-scale deposits, and composition of the environment (pH, halogens, acids, brines, and so on).

Experiences in the past will guide us in the proper alloy selection. One can either spend heavily for material initially, or count on considerable maintenance during the life of a system.

### THE ENVIRONMENTAL CHALLENGE: A MISSION POSSIBLE

It is estimated that sulphur dioxide emission will increase by three million tons in the next ten years, and probably about \$5 000-million per year will be spent on control of acid rain, clean-coal technology, wet and dry scrubbers, automotive and aircraft exhaust devices, and other emission control equipment.

A coordinated effort between the developed nations is required to clean up the atmosphere. A major step forward has been the agreement between the U.S., Canada, Japan and Western Europe to cut pollution back by another 25% over the next ten years. While not fully adequate, it is a necessary first step to be adopted by *all* nations . . . including those behind the Iron Curtain.

The industry appears to be catching its breath before beginning another period of sustained, robust growth. Much of our potential for future growth depends on our ability to come to grips with the safety and environmental challenge that confronts industry today. Nickel's contribution in air pollution abatement will help to make the mission possible!

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