

# Nickel containing materials in flue gas desulfurization equipment

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Flue gas desulfurization (FGD) is the removal of the sulfur oxides from gaseous products produced by the combustion of fossil fuels. Conventional combustion technology results in most of the sulfur in the fuel being oxidized and released into the flue gas - which contributes to the formation of "Acid Rain", a term loosely applied to the precipitation of acids from the atmosphere which can damage the environment and health. Acid Rain has become a major issue commanding the attention of governments world-wide.

Energy conservation and the burning of low sulfur fuels in place of high-sulfur fuels have played a part in achieving

reduction in total pollution, but sulfur oxide capture technology is still necessary. In this regard, current emphasis is upon removal of the sulfur oxides after combustion by wet scrubbing with alkaline slurries such as lime/limestone, with or without the recovery of gypsum.

The Nickel Development Institute (NiDI) has gained extensive experience concerning the cost-effective performance of nickel-containing materials in FGD environments. This information is available to fabricators and users of FGD equipment, materials suppliers, as well as to those providing process technology world-wide.

## Emission standards

The earliest introduction of emission standards was made on a national basis in the 1970's in Japan and then in the United States. As awareness of environmental problems increased during the 1980's, regulations became more widely applicable and stringent. This trend is continuing through the 1990's as greater understanding is gained concerning the detrimental effects of atmospheric pollution and the knowledge of what is technically and economically feasible for control of emissions.

These standards generally apply to large combustion plants (greater than 50 MWth) which include the majority of the fossil-fuel-fired boilers at electricity generating stations and other major industrial sources of pollution.

Some countries such as the United States address smaller plant emissions, with progressive tightening of legislation proving effective.

The level of regulation has varied from country to country depending upon local conditions and perceived needs. Variation in legislation has also occurred as a result of consideration of plant size, type of fuel burned and specified time limits for compliance with existing emission standards. In general, all new plants are required to meet the strictest emission limits.

### Europe

Currently, international agreement is being sought for tighter legislation under the aegis of the United States Economic Commission for Europe (UNECE) Convention on Long Range Transboundary Air Pollution.

A significant measure adopted by the European Community was the Large Combustion Plant Directive (LCPD) which defined emission limits for solid, liquid, and gaseous fuels for units exceeding 50MWth. Overall, the 26 million metric tons of SO<sub>2</sub> emitted by 14 countries were to be reduced to 15.4 million tons (14 million tonnes) by 1990 and to 9.6 million tons (8.7 million tonnes) by the year 2000, a 62% reduction.

### North America

In late 1990 the United States passed the Clean Air Act Amendment (CAAA) requiring a nationwide reduction in SO<sub>2</sub> and NO<sub>x</sub> emissions compared to the 1980 levels. The CAAA called for a two-phase program to cut SO<sub>2</sub> emission levels by 10 million tons (9.1 million tonnes) by the year 2000. After that there will be nationwide SO<sub>2</sub> emissions limit of 8.9 million tons (8.1 million tonnes) per year. By January 1, 1995, half of the total SO<sub>2</sub> emission reductions were to have been achieved (Phase 1).

By January 1, 2000, all fossil-fuel fired units over 25 MWth in size will be limited to emissions levels of 1.2 pounds of SO<sub>2</sub> per million BTU (0.5 kg per 10<sup>6</sup> joules) of heat input and will be required to have an allowance for each ton of

SO<sub>2</sub>, they emit (Phase 2). The NO<sub>x</sub> emissions reduction was originally mandated at 2000 tons per year, but the reduction is currently being renegotiated. It is estimated that US utilities will spend over 12 billion USD per year over the period 1990 to 2010 to achieve the 10 million tons (9.1 million tonnes) per year reduction in SO<sub>2</sub> emissions.

### Japan

Because of small land areas and the density of population, Japan was among the first to give considerable attention to air pollution control. In 1968, the Japanese government established the 'Air Pollution Prevention Law'. New improved technologies were developed and some of these have been licensed to North American and European companies. By the mid-1980's there were over 1500 air pollution control installations permeating all industries. Emission units are established for each specific plant in accordance with standard formulae and currently there are in excess of 15,000 MWth of electrical capacity in Japan that are equipped with scrubbers.

### Asia and the Pacific Rim

This area is experiencing rapid growth and development with increasing demand for electricity. Large quantities of fossil fuels are consumed to satisfy these requirements. Coal is increasingly utilized in India, China (including Taiwan), Indonesia, Thailand and Korea. Air quality legislation has been introduced with national emissions standards which are anticipated to match existing international levels.

## The wet scrubbing process

While FGD is essentially a simple process operating under relatively mild conditions in comparison with other chemical processes in industry, numerous material problems have been encountered.

Details and conditions under which materials function and the specific sections of FGD plants which represent problems are described in Figure 1.

Plastic and rubber linings on carbon steel are frequently specified, influenced by earlier American and Japanese experience. Failure of such linings can lead to shutdown of power stations (for example, fires in FGD units at: Rheinisch-Westfälischer Elektrizitätswerks's Neurath Plant, Grevenbroich, Germany - January 1987; Kansai Electric Power Company's Kainan Plant, Wakayama, Japan - May 1990; VEBA Kraftwerke Ruhr's Scholven Plant, Gelsenkirchen, Germany - August 1993, and in North America, Tampa Electric Big Bend Station - 1994).

The development of "wallpaper" sheet lining of carbon steel with thin (1.6 mm) (0.063 in.) nickel-containing materials, including stainless steels, is significant in this respect. Early on, it appeared that non-metallic coated carbon

steel would be the least expensive approach to the use of materials in FGD systems. However, this proved to be incorrect because the coatings are susceptible to mechanical damage and require a precise set of application conditions which if not met will result in poor adhesion. In terms of life-cycle costs and loss of revenues all-alloy construction usually offers an advantage over coated-steel systems.

There also have been failures of metallic systems resulting from improper alloy selection, poor quality welding and fabrication and insufficient knowledge or control of the service conditions. Alloy selection can now be made with confidence on the basis of experience in conjunction with a background of extensive on-site and laboratory corrosion testing.

The acid/chloride environment within an FGD scrubber can be extremely aggressive. The effects of sulfuric acid attack over a wide pH range in the presence of chlorides and fluorides derived from the fuel and accumulated in the scrubbing medium must be considered. This environment tends to initiate crevice corrosion when scaling and sludges are present.

Considerable progress has been made in the development and operation of various FGD systems. Better understanding of the process chemistry involved and of the limitations of materials of construction and of equipment design has provided improved operational performance.

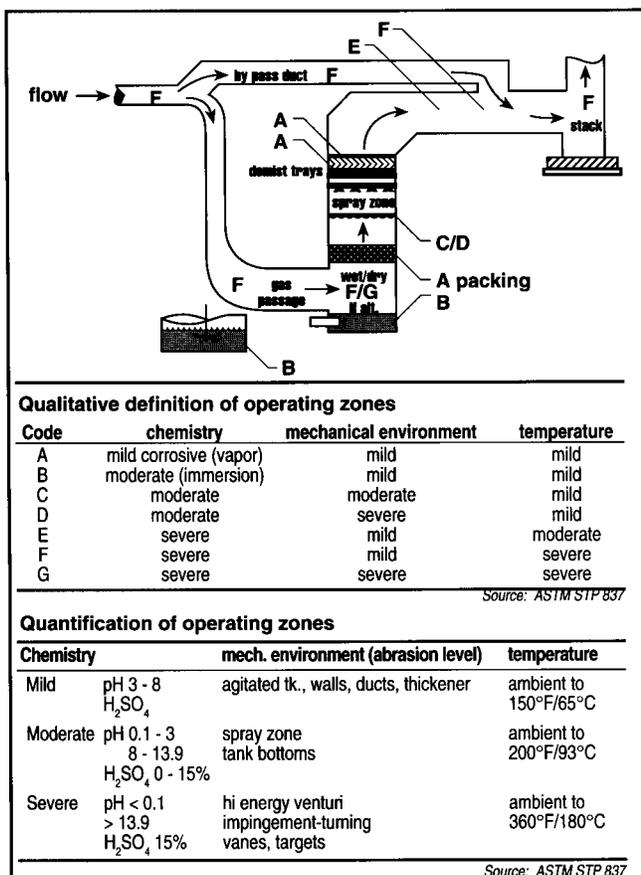


Figure 1 Operating zone generic FGD system ASTM D-33.09. (Modified)

The nickel alloys and stainless steels are, with appropriate selection and application, demonstrably able to provide cost-effective solutions to most materials problems encountered during the operation of FGD equipment. Thus, low maintenance costs along with high levels of equipment availability are insured over the plant service life expected with conventional electrical power production practices.

Extensive experience has been gained with wet lime-limestone scrubbing of flue gases, using nickel-containing materials to overcome corrosion problems. Over 40 percent of all the wet limestone/gypsum absorber towers constructed world-wide, use nickel-containing alloys. Further, it is anticipated that some 70% of all absorber towers will utilize stainless steels and nickel alloys by the year 2005.

### Factors Influencing Corrosion

#### Sulfur Content

Sulfur oxides produced from coal-combustion react with water to form corrosive acids with water. Ideally the FGD system must be able to operate with a fuel sulfur content of up to 4% with a minimum 95% removal of sulfur oxides.

#### Temperature

While the normal operating temperature range within the FGD scrubber is not high, it can have a pronounced effect on the corrosivity of the environment. Incoming flue gas is usually at approximately 160°C (320°F) and is cooled to 50 - 65°C (122 - 149°F) while passing through the scrubber system. The scrubber outlet temperature is critically close to the condensation temperature for sulfurous or sulfuric acid. Acid condensation can occur in the outlet ducts, dampers and stacks, requiring the use of acid resisting materials. It is important to appreciate that full bypass conditions with untreated flue gas may occur and must be accommodated.

From the adiabatic saturation curve (Figure 2), the approximate concentrations of sulfuric acid anticipated in condensate will range from 26 - 55% with normal operation, increasing to over 80% with full bypass. Full reheating to

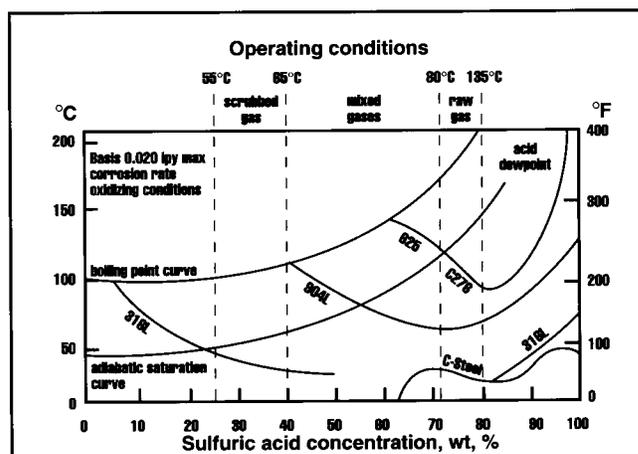


Figure 2

above 170°C (338°F) will avoid condensation at the expense of system efficiency.

Material selection can be assisted by reference to the curves in Figure 2.

#### **pH Control**

Process conditions are monitored closely to minimize scaling, in the scrubber system. Crystallization of calcium sulfite can be controlled by keeping the pH of the slurry within a suitable range. If the pH is too low, scrubber efficiency is reduced. Scrubbing slurries must generally be maintained in a slightly acid condition (4.5 - 5.5 pH) using limestone. In addition, sulfur or thiosulfate additions are increasingly used to minimize scaling. The cooling of the flue gas to optimum reaction temperatures requires water treated with neutralizing agents to avoid low pH values (1.5 or less). Optimum temperatures differ as determined by the fuel used; for example, they are higher with brown coals (Lignite) such as are used in Europe. These are operating constraints which must be considered in the selection of materials of construction.

#### **Chloride Content**

A further factor concerning corrosivity of the scrubber environment is the presence of chlorides from the scrubber water and also from the absorption of hydrogen chloride generated by the combustion of chloride containing coals. Chlorides can concentrate because of evaporation and recirculation of water, in single loop circulation systems to levels reportedly exceeding 100,000 ppm resulting in highly corrosive acid chloride solutions. This is important with the imposition of strict controls on chloride discharge in waste waters. Methods such as multi-stage flash distillation or blow-down may be necessary for chloride removal from process water discharged from the FGD plant.

In a two-loop system the high chloride environment can be restricted to the quench area of a scrubber. The remaining scrubber areas with chloride contents in the few thousand ppm range will encounter less corrosive conditions and may not require the most resistant nickel alloys.

#### **Fluoride Content**

Up to 3,000 ppm of fluorides may be found in coals. Fluorides tend to concentrate on metal surfaces under scale deposits aggravating acid-chloride crevice corrosion of the stainless steels. A nickel-base alloy, with higher chromium and molybdenum contents, may be required to resist these corrosive effects from chloride and fluoride levels which may reach 100,000 ppm or more.

## **Corrosion resistance of nickel materials**

Stainless steels and nickel alloys owe their corrosion resistance to a thin, passive surface-oxide film, rich in chromium, which spontaneously forms when exposed to air. This film-forming property is common to all alloys containing more

than approximately 12% chromium.

The composition of the oxide film obviously varies with the composition of the material. However even if the film breaks down, it can still provide resistance to corrosion in many environments.

For almost every application there is a grade of material which will offer satisfactory and cost-effective service.

## **Corrosion in FGD equipment**

### **Stress-Corrosion Cracking**

Stress-corrosion cracking of the austenitic stainless steels is not a problem at the normal operating temperatures of scrubbers, 50 - 65 °C (122 - 149 °F), but may become a problem at temperatures of 150 - 175 °C (302 - 347 °F) and higher.

It is important to recognize that even when pH and chlorides of the bulk solution are very well controlled, there is always the possibility of concentration of chlorides under deposits or on heat-transfer surfaces that can lead to the conditions required for stress-corrosion cracking.

### **Pitting Effect of Chloride Concentration and pH**

There has been extensive research into the influence of chloride-ion concentration, pH and temperature on the performance of nickel-containing materials in process industry applications and with the handling of sea-waters. Much of this is relevant to the conditions experienced in FGD equipment. There is a definite correlation between localized corrosive attack and the pH and chloride levels. Deposited solids can lead to localized attack by concentrating chlorides and by providing oxygen-deprived crevice conditions. Materials selection must take into account the likelihood of such deposits in operation.

Tests under controlled conditions have helped to define the pH/chloride limits for various nickel alloy materials of construction. This information can be used to indicate conditions where the stainless steels and the more corrosion resistant nickel alloys are required.

The selection of appropriate alloys to satisfy conditions encountered in FGD equipment, can be facilitated by the use of the Pitting Resistance Equivalent Number, (PREN) which can be used to compare materials, based upon the formula:

$$\text{PREN} = \%Cr + 3.3x\%Mo + 16x\%N$$

When tungsten is present, the following formula may be used:

$$\text{PREN}_w = \%Cr + 3.3x\%Mo + 1.65x\%W + 16x\%N$$

The higher the number, the more resistant the alloy is to pitting attack. The PREN represents an extension of the pitting

index used successfully with stainless steels in seawater.

The performance of Type 316L, Type 317L and Type 317LM stainless steels is plotted in *Figure 3*. It provides a conservative guide to alloy selection for FGD scrubbing systems in the range 3.5 - 7.0 pH and up to about 5,000 ppm chlorides and demonstrates the use of the PREN.

**Effect of Temperature**

In general, an increase in temperature increases the rate of corrosion attack or degree of susceptibility of the alloy to initiation of corrosion. To accommodate higher temperature operation in FGD equipment or to take into account the possibility of temperature excursions in service, higher grades of nickel-containing materials capable of resisting the more severe conditions are available.

**The Influence of Molybdenum**

It is known that molybdenum enhances the resistance of austenitic stainless steels and nickel alloys to general corrosion, and especially to pitting in chloride-containing solutions and sulfur dioxide vapors. For example the recognition of the value of increased amounts of molybdenum has led to the development of Type 317LM stainless steel with molybdenum contents of 4.0 to 4.5%Mo exceeding the normal specification range for 317 stainless steel for certain corrosive applications. Too, the 6 %Mo super austenitic stainless steels with even higher molybdenum contents have been developed to provide greater resistance to acidic chloride environments.

**Nickel materials - application in FGD equipment**

Much of the early FGD equipment was designed around the use of carbon steel components with non-metallic coatings and rubber linings because of a lack of understanding of the conditions to be encountered. Too, there was a lack of reliable data concerning the performance of stainless steels and nickel alloys. Despite considerable progress with improved coating technology, problems remain which may be resolved by the selection of an appropriate grade of nickel containing alloy.

Corrosion resistance is improved by increasing chromium, nickel, molybdenum and nitrogen contents. Chromium provides passive protective surface films and molybdenum and nitrogen improve the resistance to pitting and crevice corrosion. Nickel assists in renewing damaged protective films and also improves fabrication and weldability.

The problem facing designers is to specify the material that contains the appropriate combination of these alloying elements which will provide resistance to corrosion in FGD scrubber environments at least cost.

**Use of the Stainless Steels**

Standard stainless steels are specified where applicable, but increasingly, stainless steels containing higher molybdenum, chromium and nickel contents are being selected because more severe conditions are being encountered. Low carbon or titanium or niobium stabilized grades of stainless steel provide resistance to intergranular corrosion. The problem of chloride stress-corrosion cracking (CSCC) can be resolved, for example, by specifying alloys containing in excess of 42% nickel or having an austenitic-ferritic microstructure such as the 22% and 25 %Cr duplex alloys.

**The Use of Nickel-Chromium-Molybdenum Alloys**

It has been determined that the modified grades of stainless steels such as Type 317LM or the nitrogen-containing Type 317LMN will have inadequate corrosion resistance in scrubber environments with a combination of high chloride levels and low pH. Utilizing the Pitting Resistance Equivalent Number (PREN) for guidance and the results of practical experience backed by extensive laboratory testing, a performance ranking of the nickel stainless steels and the higher nickel-chromium-molybdenum alloys has been developed. Its purpose is to assist in the selection of the most effective material to resist specific corrosive conditions involving decreasing pH values and increasing chloride levels.

The guidelines established consider conditions of varying corrosion severity at normal operating temperatures (50 – 65 °C). It is important to appreciate that the effects of temperature deviations, the presence of fluorides, heavy scaling, and undesirable features in design and fabrication

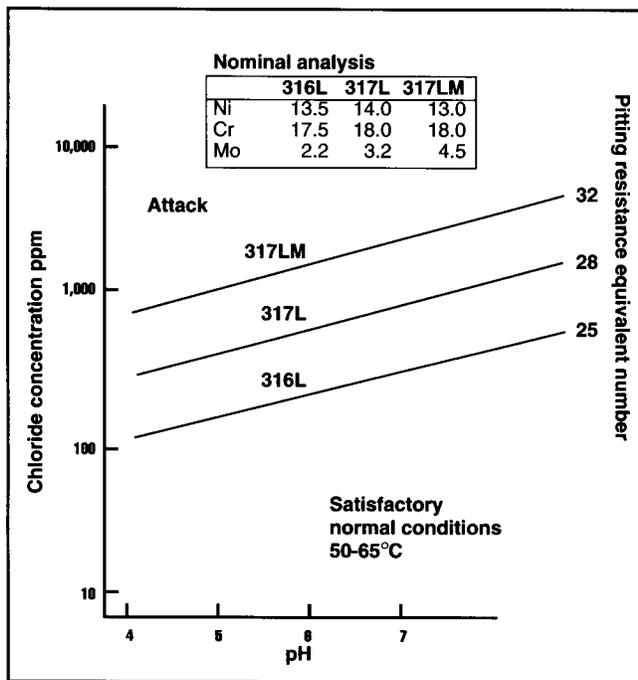


Figure 3 Limits of use of stainless steels FGD scrubbers (Modified Schillmoller diagram).

can influence alloy behavior and cannot be interpreted in this simplified way.

It is also well to keep in mind that the environment in each scrubber is unique no matter how similar the design is to that of another. The fuel combusted, the water utilized, the operation of the unit and slight differences in unit design can affect the performance of materials.

Figure 4 suggests alloy selection guidelines for chloride concentrations up to 200,000 ppm and pH values from 1 - 6.5. To facilitate consideration of the detail provided, the bases "mild" to "very severe" conditions of pH and chloride concentrations are indicated.

Similar data considering the influence of fluorides together with chlorides have been published.<sup>1</sup>

The nominal compositions of the nickel materials referred to in Figure 4 are given in *Table I*.

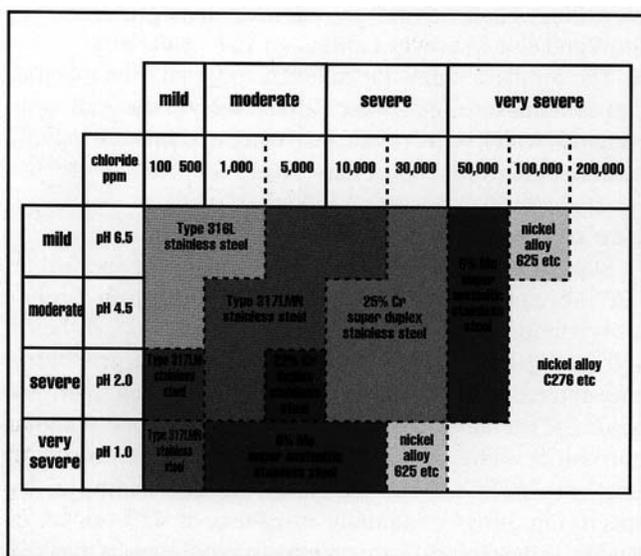


Figure 4 Guideline stainless steel and nickel alloy selection for FGD equipment.

## Minimizing problems

There are several factors in the design and operation of alloy FGD scrubbers that will help minimize or prevent problems with corrosion to give long, trouble-free service life:

- Monitor chloride levels and pH ranges
- Prevent chloride concentration
- Avoid crevices in design and construction
- Keep surface free of slurry accumulation or scale
- Specify appropriate alloys according to operating conditions within the scrubber.

Monitoring chlorides in itself will not prevent corrosion but will give a good indication of potential trouble. Whenever there is recirculation, there is a potential for concentrating chlorides to aggressive levels. Control of pH is essential for scrubbing efficiency and to prevent scaling. A significant reduction in pH can lead to accelerated corrosion.

Preventing chloride concentration is primarily a matter of process design. If chlorides concentrate, it is best to confine that concentration to a small area of the system if possible and to specify materials in that area that will resist attack at the highest chloride levels anticipated.

Avoiding crevices in the design and construction of any chemical process equipment is always good practice, particularly where condensate can collect and concentrate to a much higher acid level than expected.

Preventing condensation is important because scrubber flue gas condensates may be aggressive. If reheaters are used, it is important that heat should be maintained within the tube bundle during shutdowns to prevent condensation. It should be noted that most current FGD designs do not include reheaters.

Keeping equipment free of deposits is recommended. Accumulation of deposits and scale increases the risk of pitting and crevice corrosion. Thus, cleaning during shutdowns should also be done.

Table I Compositions of representative nickel-containing materials utilized for FGD equipment

Material	Nominal Composition	- Ni	- Mo	- Cr	- Cu	- W	Weight Percent Nb	Weight Percent N	Pitting Resis. Equiv. PREN <sub>w</sub>
Type 316L	14.5	>2.75	18	-	-	-	-	0.02	27
Type 317LM	15.5	4	18	-	-	-	-	0.06	32
Type 317LMN	14.0	>4.5	18	-	-	-	-	0.16	33
Duplex	5.5	3	22	-	-	-	-	0.17	34
Super Duplex	6.5	3	25	-	-	-	-	0.22	38
Super Aust.	25	6.5	21	1	-	-	-	0.22	46
Alloy N06625	61	9	22	-	-	4	-	-	51
Alloy N06020	59	13	22	-	3	-	-	-	70
Alloy N10276	56	16	16	-	3.5	-	-	-	75
Alloy N06686	57	16	21	-	3.9	-	-	-	80

## Fabrication

Nickel materials are often disregarded during consideration of material specification for equipment because of the high cost of alloys compared to other materials in terms of cost per unit weight. However, effective reduction of the cost of equipment has been achieved by utilizing one or more of the following:

- light weight structural design
- structural grade steels pre-clad with nickel materials
- "Wallpaper" (sheet) lining of structural steel equipment with nickel-containing materials

As an indication of cost reductions achievable, a comparison of relative costs for non-metallic and metallic systems is illustrated in *Table II* for FGD absorbers.

### "Wallpapering" (Sheet Lining)

Considerable emphasis is being placed upon the "wallpapering" technique, using 0.063 in. (1.6 mm) sheets of alloy. Appropriate sheet widths and lengths are chosen to permit convenience in handling and the least amount of welding.

*Figure 6* illustrates the overlap technique employed to ensure attachment to the substrate and total sealing of the alloy sheets. Plug or arc-spot welds at suitable intervals (usually 2-foot centers) satisfactorily minimizes vibration.

**Table II** Detailed 1997 Cost Comparison.<sup>2</sup>  
The following absorber cost comparisons are based on a 0.25 inch thick plate knockdown construction of a standard design.

Material	UNS	Procurement Fabrication	Erection	Total
A-36 Carbon Steel (CS)	K02600	1.00	1.00	1.00
CS Elastomer-Lined		5.79	1.00	2.26
CS Flakelglass-Lined		5.36	1.00	2.15
316L Stainless Steel	S31603	4.00	1.00	1.79
317L Stainless Steel	S31703	4.40	1.00	1.89
317LMN Stainless	S31726	5.30	1.00	2.13
Super Duplex Stainless	S32550	7.00	1.05	2.58
6% Mo Alloys	N08367/S3125	7.50	1.05	2.71
Nickel Alloy	N06022	14.00	1.19	4.55
Nickel Alloy	N10276	14.00	1.19	4.55
Nickel Alloy	N06686	15.10	1.19	4.84
Nickel Alloy	N06059	14.83	1.19	4.77
CS w/Alloy N06022	Cladding	9.50	1.14	3.34
CS w/Alloy N06022	Wallpaper	4.50	1.43	2.30
CS w/Tile Lining		6.20	1.00	2.37
Concrete Block		10.00	Included	2.63

Note: Costs of all options are presented relative to the base cost of 1.0

## Lining technique

The wallpapering (sheet lining) technique is well established, having been employed in the chemical and process industries for over 30 years. Too, experience has been developed with FGD equipment in the United States and Europe for more than 14 years.

Concern has been expressed regarding the spot or plug welding weld quality with the possibility of weld dilution from the structural steel. The latter problem has been eliminated in the case of plug welds with the use of alloy caps welded over the plug welds. With the arc spot welds, the continuous capping of the weld deposits with filler metal as the process is completed eliminates weld dilution. Thermal stressing associated with differential thermal expansion between structural steels and nickel materials has also been questioned. Tests and practical experience have shown that the latter do not present problems at the temperatures encountered. Weld parameters can be closely defined and well monitored with electronic equipment utilized in conjunction with pulsed arc GMA (MIG) welding machines.

Wallpapering or sheet lining of FGD equipment using microprocessor controlled, pulsed arc GMA welding equipment is a relatively simple operation that permits cost-effective use of the lining materials. However, wallpapering using the GMAW short circuiting technique must be avoided because of the poor quality welds that are produced.

## Life cycle costing

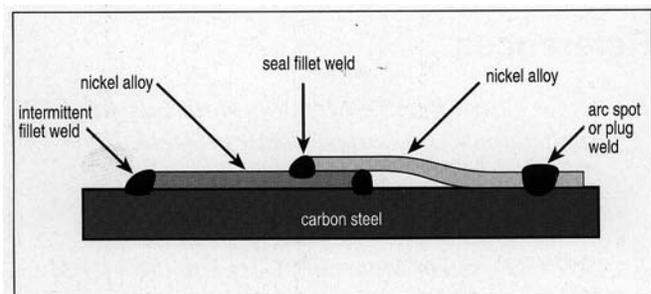
Life-Cycle cost comparisons have been developed for a range of materials utilized in scrubbers. Specific detail is incorporated in the Report published by NiDI entitled:

"Life-Cycle Cost Benefits of Constructing an FGD System with Selected Stainless Steels and Nickel-Base Alloys".

The comparison covers a combined FGD scrubber system comprised of a solid Alloy C-276 venturi and Alloy C-296 clad absorber and Alloy C-276 clad ductwork.

The results show that even with the most costly of the nickel alloys considered the life-cycle costs are lower than that of a similar system using Neoprene rubber-lined carbon steel.

(*Figure 7*)



**Figure 6** Lining technique.

Alloy C-276 may not be an essential requirement and different alloys may be used in different areas of the same equipment - further reducing actual costs in line with the data shown. Too, the use of sheet linings, wallpapering may reduce the costs even further. Thus, it can be determined that nickel-containing materials offer cost-effective solutions to corrosion problems in FGD equipment with low maintenance costs and high equipment availability over the life of the facility.

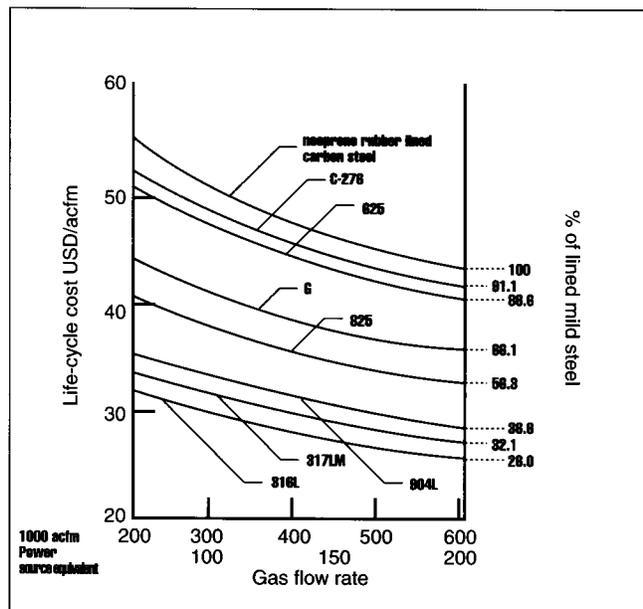


Figure 7 Life-cycle cost comparison, combined system.

## Conclusion

### Advantages of Nickel-Containing Materials

It should be recognized that the corrosion resistance of nickel-containing materials is an inherent quality. Generally, the nickel-chromium alloys with molybdenum and other alloying elements provide a stable passive film that protects the alloy against corrosion by the severe environments frequently encountered in FGD systems.

The elevated temperature strength of the nickel-containing materials specified for FGD scrubbers ensures structural integrity should the temperature exceed design expectations. This eliminates the need for any backup cooling water systems that could be required with non-metallic systems.

The erosion resistance of nickel-containing materials is adequate for moderately abrasive slurries, especially in corrosive environments.<sup>4</sup> In a wet system, erosion and corrosion usually occur together, not as two separate phenomena.

The good mechanical properties of nickel-containing materials can be used to advantage in scrubber construction. Design of vessels can incorporate the use of thin-gauge materials with exterior reinforcement.

The fabrication characteristics of nickel-containing materials are excellent. These materials can be cut, formed and joined by methods similar to those used for carbon steel fabrication, although care must be exercised to preserve the inherent corrosion resistance. Too, shop fabrication can reduce costs below those of site fabrication, contributing further to the economy of the installation.

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