

Flue-gas sulfur-recovery plant for a multifuel boiler

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ABSTRACT: In October 1991, a Finnish fluting mill brought on stream a flue-gas desulfurization plant with an SO₂ reduction capacity of 99%. The desulfurization plant enabled the mill to discontinue the use of its sulfur burner for SO₂ production. The required makeup sulfur is now obtained in the form of sulfuric acid used by the acetic acid plant, which operates in conjunction with the evaporating plant. The mill's sulfur consumption has decreased by about 6000 tons/year (13.2 million lb/year) because of sulfur recycling.

KEYWORDS: Boilers, chemical recovery, emission, flue gas, sulfur compounds, sulfur dioxide.

The Savon Sellu fluting mill in Kuopio, central Finland, was started up in 1968. Today it produces 225,000 tons/year of fluting board. Because of its ammonia-based cooking process, the mill was one of the largest sulfur dioxide emission sources in Finland. Maximum annual emissions were on the order of 14,300 tons SO₂/year.

In 1989 the regulatory authorities passed an air-pollution control resolution limiting the mill's sulfur emissions to a maximum of 15 kg SO₂/ton of NSSC (neutral sulfite semichemical) pulp from January 1, 1993. In effect, this meant a reduction of 75% in emission levels.

The mill immediately began evaluating the various purification methods available. Based on considerations of

scheduling, scrubber experience, cost, and energy use, the mill selected Tampella Power to provide a complete turnkey desulfurization system. The recovery-process system cost about FIM 30 million (US\$ 4.5 million).

Process

The primary goal underlying the design of the desulfurization plant was to create a closed sulfur cycle that met the emission limits imposed by the environmental authorities. At the same time, provision was made for tighter air-pollution controls in the future. A second goal was to shut down the mill's old cooking-liquor production plant.

Environmental investment in an old boiler

The mill's main boiler was a multifuel unit—120 tons/h (265,500 lb/h); 525°C (975°F); 115 bar (1665 psig)—placed in use in 1968. The boiler burns the mill's spent liquor, peat, bark, and heavy fuel oil. The boiler is equipped with a peat-drying plant and an electrostatic precipitator (ESP) for removing particulates from the flue gas. The spent pulping liquor flow into the boiler at 18 m³/h has a dry-solids content of 55% and a sulfur content of 8.5%.

The new air-pollution control solution required that the cooking method be unaffected by the installation of the new desulfurization plant. This, in turn, led to the adoption of an unconventional approach to the sulfur-recovery process. Recovery takes place from the cooled boiler flue gases and at high flow, with a high dust content and a low sulfur dioxide concentration. The desulfurization system also cleans the flue gases (mostly of SO₂) produced by an oil-fired auxiliary boiler. Table I gives the process design data. Figure 1 is a flowsheet of the pulp and board mill.

Multistage process for SO_x recovery

The new desulfurization process was designed around a spray-wash scrubber. The supplier had previously delivered more than 30 such units for sodium-based sulfite processes in Scandinavia. These units had shown excellent results in the removal of sulfur and particulate. The properties of the flue-gas flow led to the design shown in Fig. 2. Since the process was

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Flue-Gas Desulfurization

the first of its kind, the designers chose to make the plant multistage to provide a variety of different operating modes. Generous provision was also made for future development.

In the first stage of the process, remaining particulate matter in the flue gases is removed in the spray-wash scrubber either by water or condensates from the evaporator. In the second stage, the flue gases are passed through a direct-contact heat exchanger to reduce their temperature to the optimum suitable for the three absorption stages. The heat recovered at this stage is used to provide hot water for the mill and to preheat the combustion air. SO₂ absorption at the washing and cooling stages is prevented by maintaining the pH below 3. To do this, however, it is necessary to use special materials, such as fiber-glass-reinforced plastics and acid-resistant steels.

In the final three stages—absorption—the SO₂ in the flue gas is absorbed into secondary-condensate-based ammonium water, producing a liquid of ammonium sulfite, which is used as cooking liquor without further treatment. Secondary condensate is used to minimize the use of fresh water.

The absorption stages are based on packed-bed technology, which has lower energy consumption because the pressure loss over the scrubber is lower than most other scrubber technologies. Also mass-transfer properties and serviceability are good. The SO₂ content of the cooking liquor is regulated according to the results given by an automatic titrator. The SO₂/NH₃ ratio is controlled by the pH control circuit.

Trial runs

The plant was taken into operation in October 1991, one month earlier than agreed, and well over one year before the deadline stipulated by the authorities. This allowed time to test and fine tune the plant, which was desirable in view of the prototype process adopted.

Trial runs began after a short testing period. During the trial run period,

I: Desulfurization process specifications. Flue gas characteristics after electrostatic precipitator (ESP)(design values).

Gas flow, m ³ n/s	W ^b	56	SCF	1980
Temperature, °C		140	°F	285
Pressure, Pa		-150	psig	-0.02
SO ₂ content, mg/m ³ n	D ^c	9800	ppm	3430
SO ₃ content, mg/m ³ n	D	300	ppm	85
O ₂ content, % -vol		3.6		
CO ₂ content, % -vol	D	11.2		
N ₂ content, % -vol	D	62.4		
H ₂ O content, % -vol		22.5		
Particulates, g/m ³ n	D	0.400	gr/SCF	0.17
NH ₃ solution to scrubber				
NH ₃ content, %-weight		18		
Temperature, °C		30	°F	85
Pressure, MPa		0.3		

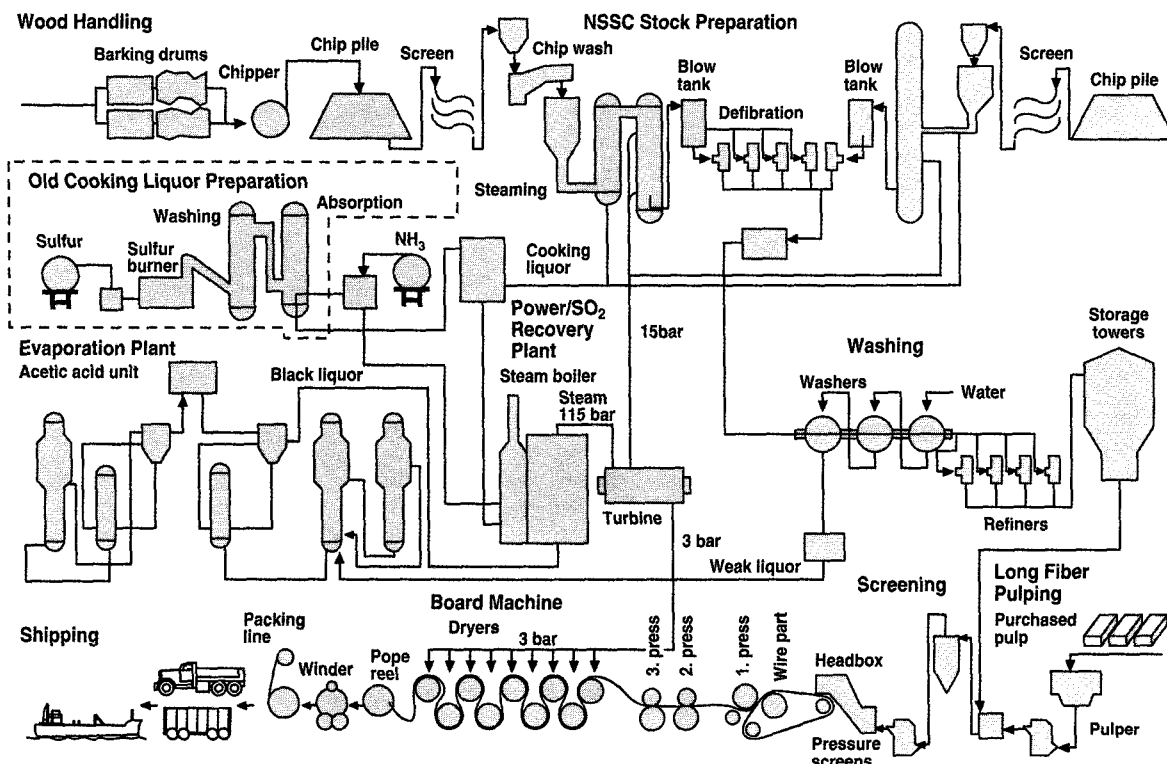
^am³n = Nm³(NTP)
^bW = wet basis
^cD = dry basis

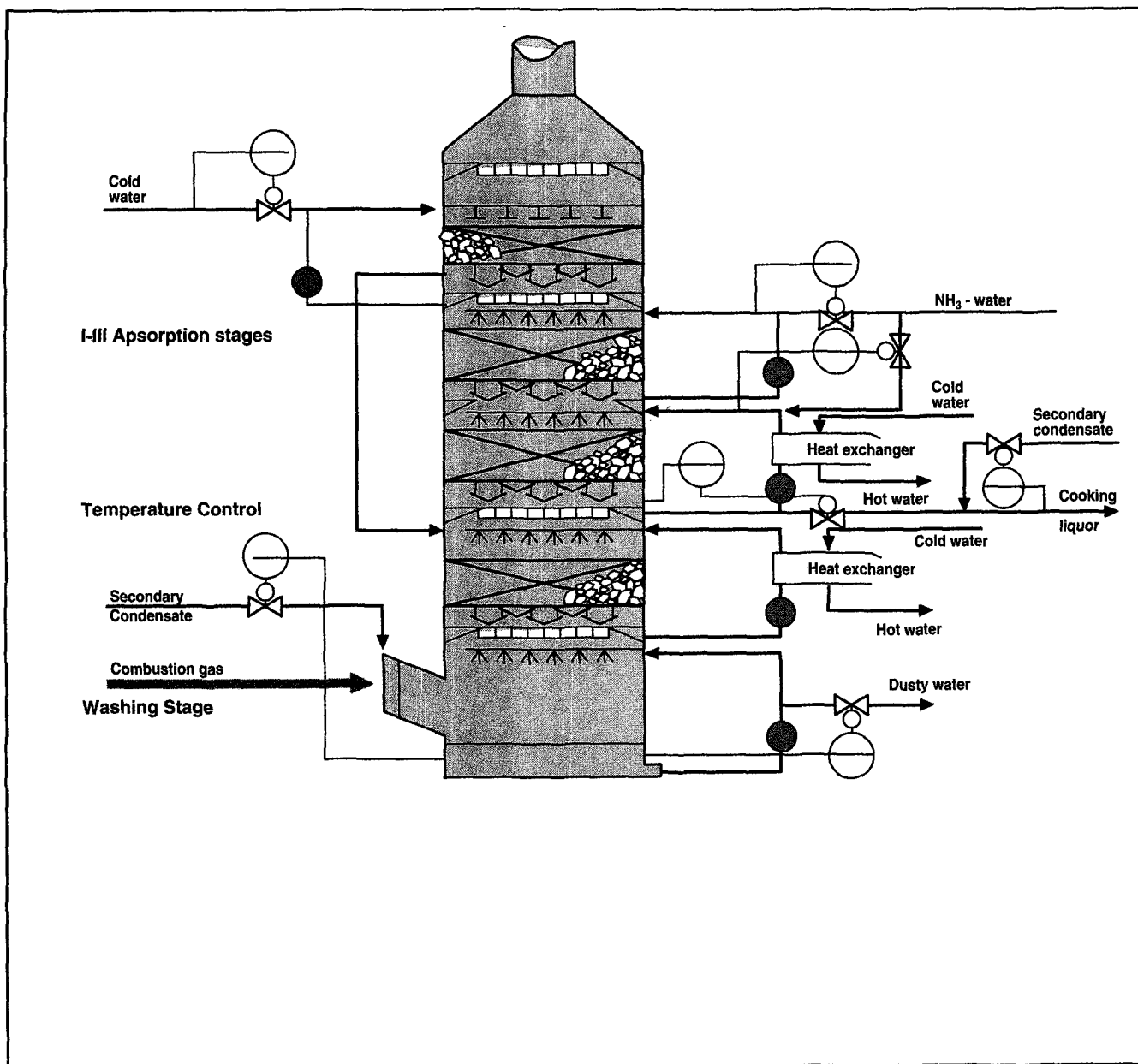
II: Results of startup tests

	Oct. 10, '91	Oct. 11, '91
Absorption operating model		
pH	5.8	5.9
Temperature, °C (°F)	51 (124)	67 (153)
Flue gas		
Before plant		
* O ₂ , %-vol W ^b	4.5–5.5	4.5–5.0
* flow, m ³ n/s (SCF/s)W	63–65 (2200–2300)	61–65 (2150–2300)
* SO ₂ , mg/m ³ n (ppm) D ^c	9500–14,400 (3300–5000)	12,800– 13,800 (4500–4800)
After plant		
* SO ₂ , mg/m ³ n (ppm) D	160–290 (55–100)	105–150 (35–50)
* NH ₃ , mg/m ³ n (ppm) D	10–15 (15–20)	200 (260)
* Particulates, mg/m ³ n (g/SCF) D	-	10–15 (0.004–0.006)
Cooking liquor		
* SO ₂ , %-weight	66–69	62–67
* Dry solids, g/m ³ (g/ft ³)	45 (19.5)	40 (17.5)

^am³n = Nm³(NTP)
^bW = wet basis
^cD = dry basis

1. Flowsheet for the pulp and board mill





special attention was paid to fine tuning the process values since, right from startup, the plant was producing suitable cooking liquor for the mill's pulp process.

Following the trial runs, tests were made to assess the effect of the temperature of the absorption stages on NH_3 emissions. At the same time, the operation of the dust-washing stage was tested at different gas flows and with different washing solutions. In light of the test results, the absorption-stage values (pH, temperature)

were set to give an SO_2 content of the cooking liquor of 55 g/L, as required by the mill, and to minimize SO_2 and NH_3 emissions (Table II).

During the tests, the liquid circulation of the flue-gas washing stage had a dry-solids content of 400–450 mg/L. The liquid at the cooling stage had a dry-solids content below 20 mg/L, indicating that dry-solids recovery from the flue gas was extremely high. The temperature of the absorption stage had a direct effect on the NH_3 emissions, which are similar to the labora-

tory and theoretical studies made before designing of the recovery plant.

Guarantee measurements

The measurements were carried out as a team effort by the mill, the supplier, and an outside consultant.

The SO_2 and NO_x contents of the flue gas were measured before and after the plant using on-line analyzers. On-line measurements were also taken of the O_2 , CO , CO_2 , and TRS

III: Results of guarantee measurements

	Dec. 3, 1991	Dec. 4, 1991
Main boiler		
Steam production, ton (lb/h)	125 (276,500)	125 (276,500)
Fuels		
* Spent pulping liquor, m ³ /h (gal/min)	13 (57)	15 (66)
* Peat feed, %	30-50	40-70
* Heavy fuel oil, ton/h (gal/min)	0.8-1.6 (3.8-7.5)	1.3-1.6 (6.1-7.5)
* Bark, m ³ /h (dry tons/h)	9-12 (3-4)	6-9 (2-3)
Desulfurization plant		
Absorption section		
* pH	6.5	6-6.5
* Temperature, °C(°F)	57 (135)	57 (135)
* Cooking liquor flow, m ³ /h (gal/min)	34 (150)	35-40 (155-175)
SO ₂ , %-weight	5.6	5.6
Flue gas		
Before plant		
* Temp., °C(°F)	135-150 (275-300)	145-150 (295-300)
* O ₂ , %-vol, W	4.5-6.0	4.0-5.0
* CO ₂ , %-vol, D	14.3-15.5	12.5-13.0
* CO, ppm, D	160-760	1500-2030
* SO ₂ , mg/m ³ n (ppm)	10,300-12,100 (3600-4230)	10,800-11,900 (3780-4160)
* NO _x , mg/NO ₂ /m ³ n (ppm)	275-445 (135-215)	210-320 (100-155)
* TRS*, mg H ₂ S/m ³ n (ppm)		
D	0	10-65 (7-45)
* Flow, m ³ n/s (scfm)		
W	65-68 (136,500-144,000)	60-67 (126,000-140,700)
* Humidity, %-vol	22-28	19-24
After plant		
* SO ₂ , mg/m ³ n (ppm)		
D	70-100 (25-35)	130-190 (45-65)
* NO _x , mg/NO ₂ /m ³ n (ppm)	220-380 (110-185)	195-330 (95-160)
NH ₃ , mg NH ₃ /m ³ n (ppm)		
D	10-20 (15-25)	10-20 (15-25)
TRS, mg H ₂ S/m ³ n (ppm)		
D	0	10-45 (7-30)
*TRS = total reduced sulfur		

(total reduced sulfur) concentrations before the plant.

The NH₃ content of the flue gas was determined by phased wet method (sampling into 1% HCl, analysis by ammonium ion-selective electrode).

During the guarantee tests, the operation of the plant was tested to establish whether the set guarantee values were achieved in practice. The results of these tests are presented in **Table III** together with certain operating values.

The SO₂ value measured after the plant met the guaranteed value given for the plant of 400 mg/m³. Similarly, the NH₃ value complied with the guaranteed values [20 mg NH₃/m³n (25 ppm), dry gas] when the plant was operated according to the terms specified in the guarantee. **Figure 3** shows the SO₂ emissions in the flue gases measured at the plant, while **Fig. 4** gives the NO_x emissions measured during the guarantee tests.

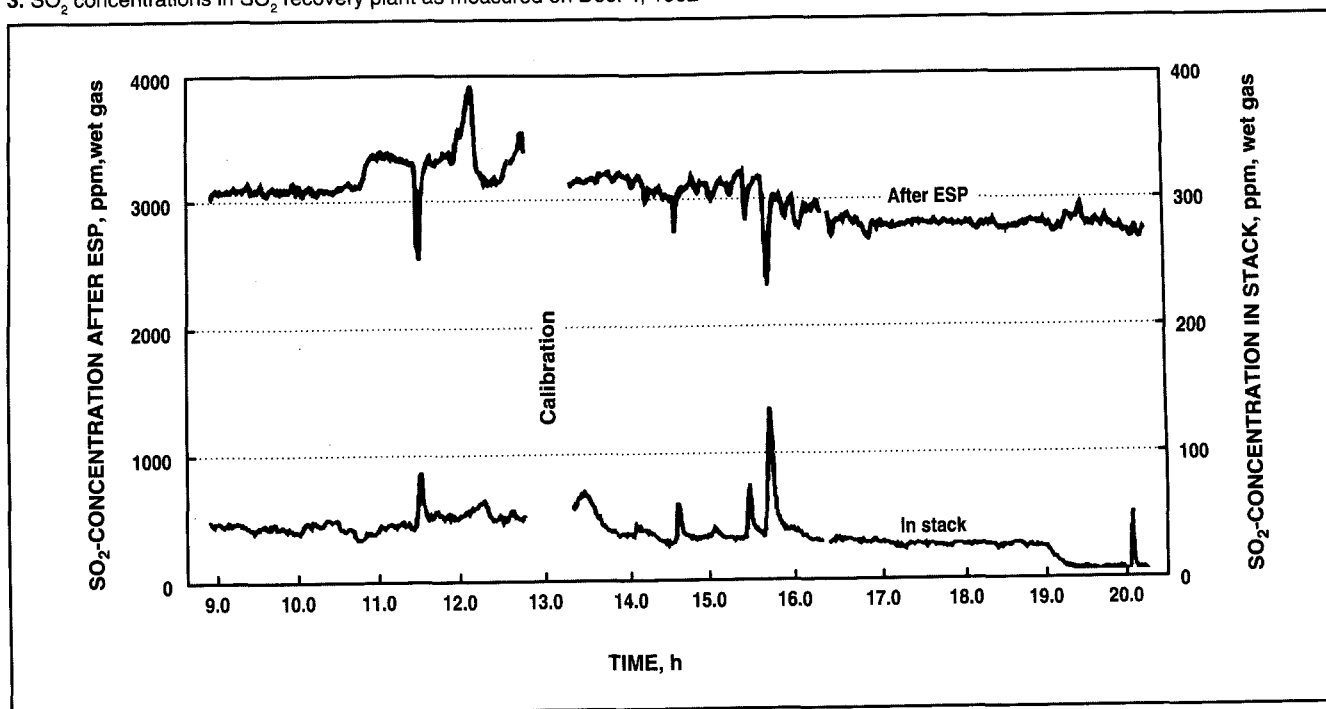
The outside consultant took samples of the flue gas before and after the plant. A chromatograph was used to analyze the different sulfur compounds in these samples as well as various volatile compounds. The analyses showed that the sulfur compounds in the flue gases were SO₂ and H₂S compounds. The degree of SO₂ recovery measured was over 99%.

Operating experience

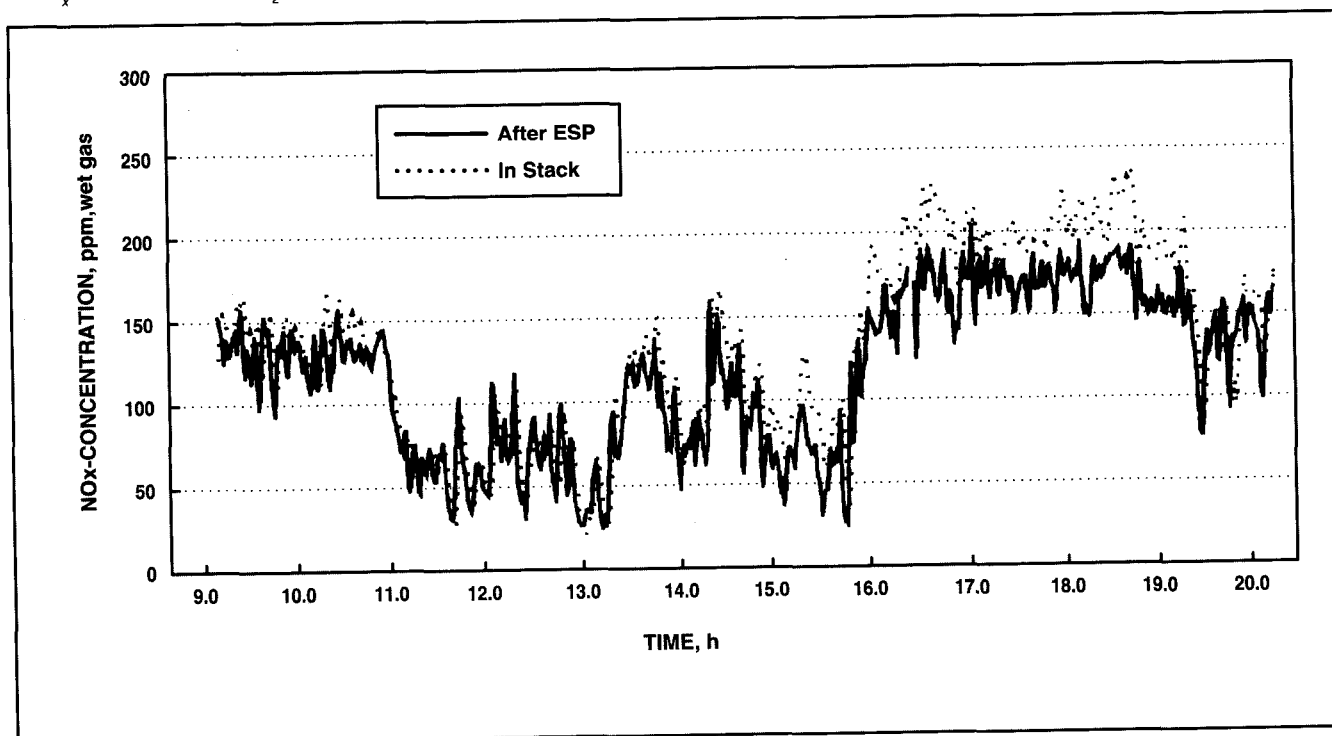
The desulfurization plant has been in production operation since October 1991. Its availability has been 100%. The level of SO₂ reduction currently required by the authorities is 75%. However, the plant achieves an SO₂ reduction of 99%, which is expected to meet the most stringent regulations likely to be passed in the future. Particulate emissions are below 10 mg/m³n.

The introduction of the desulfurization plant also enabled the mill to discontinue the use of its sulfur burner for SO₂ production. The required makeup sulfur is now obtained in the form of sulfuric acid used by the acetic

3. SO₂ concentrations in SO₂ recovery plant as measured on Dec. 4, 1992



4. NO_x concentrations in SO₂ recovery plant as measured on Dec. 4, 1992



acid plant, which operates in conjunction with the evaporating plant. The mill's sulfur consumption has decreased by about 6000 tons/year (13.2

million lb/year) because of sulfur recycling.

Experience so far indicates that the novel solutions adopted by the mill have been successful.

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