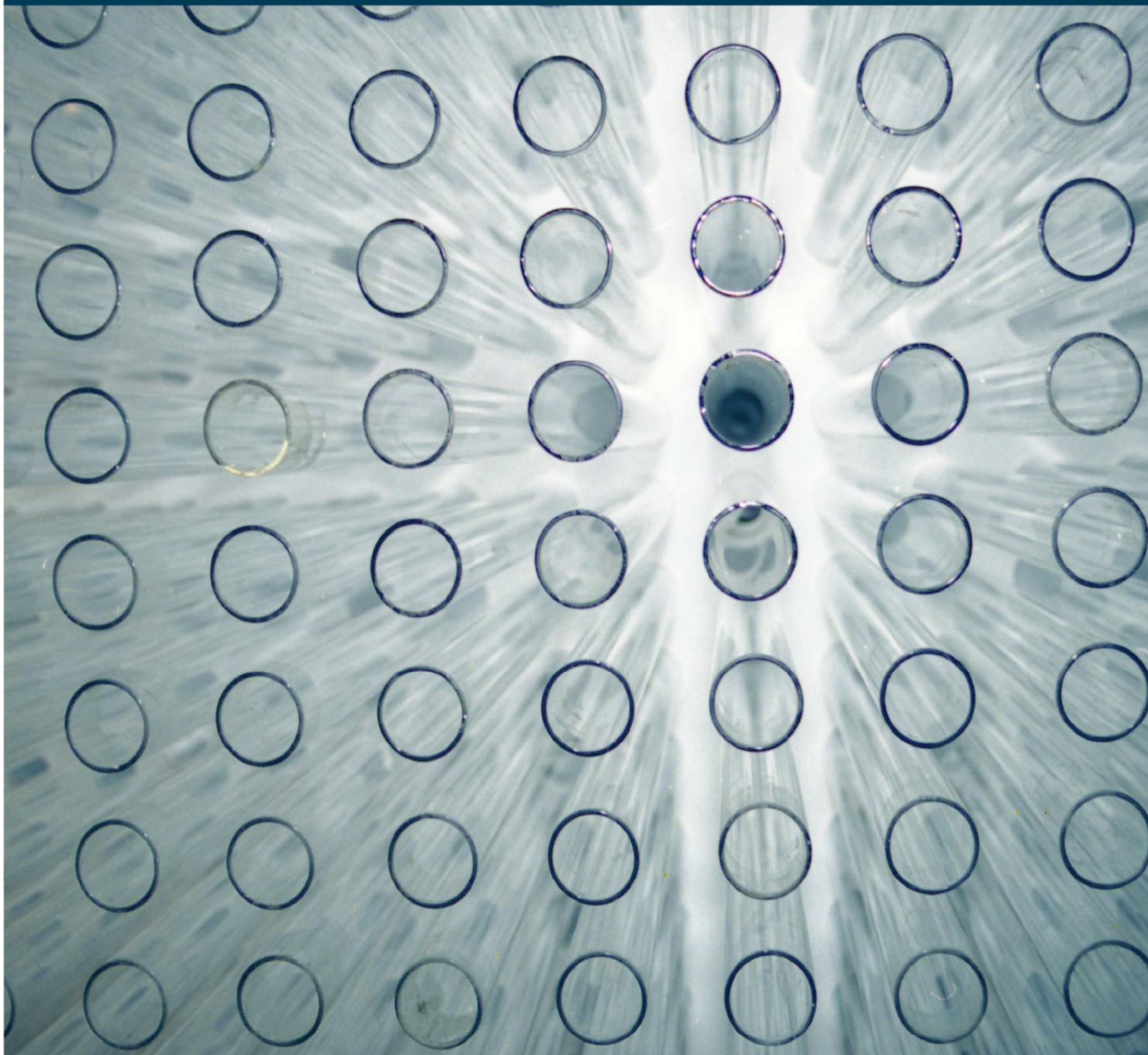


SNOX™ flue gas treatment for boilers burning petcoke ...makes petcoke more attractive for power and heat generation

RESEARCH | TECHNOLOGY | CATALYSTS



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Summary

The SNOX™ process is a regenerative, catalytic flue gas cleaning process, which removes up to 99% of SO₂ and SO₃, up to 96% of NO_x and essentially all particulates from flue gases. The sulfur is recovered as commercial grade concentrated sulfuric acid while NO_x is reduced to N₂. The process does not consume water or materials, except for ammonia for the catalytic NO_x reduction, and it does not generate any secondary sources of pollution, such as waste water, slurries or solids. The operation cost of a SNOX™ plant decreases with increasing sulfur content in the flue gas, even before taking credit for sales of the produced sulfuric acid. The process is in particular suited for treatment of flue gases with high contents of SO₂ and SO₃ and heavy metal oxides from combustion of petroleum coke and other petroleum residues with none of the environmental and operational problems encountered with other FGD processes. With SNOX™-equipped downshot boilers, up to 100% high sulfur petcoke can be burned with no more environmental or operating problems than with normal coal fired boilers equipped with normal FGD scrubbers.

Today, the SNOX™ process is used in large scale on a coal fired power plant in Denmark, a petcoke fired power plant in Italy and soon in a heavy residual fuel oil fired steam and power plant in Austria. The paper describes design and operating experience of the 1,200,000 Nm³/h SNOX™ plant at the petcoke fueled power plant of the Gela Refinery in Gela, Sicily, Italy and compares SNOX™-equipped downshot PC boilers with alternative processes for combustion of petcoke in power stations. The SNOX™ plant in Gela has operated with more than 98% availability and practically unchanged performance since it was commissioned in September 1999.



1,200,000 Nm³/h SNOX™ plant at the power plant of Raffineria di Gela, Italy



300 MW SNOX™-equipped power plant of Nordjyllandsvaerket, Denmark

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Introduction

More and more heavy crude oil with high content of non-volatile carbon and high sulfur and metal content is processed, and more and more process units are introduced in the oil refineries in order to squeeze out of the crude oil as much valuable distillate product as possible.

Delayed coking is the least expensive way of converting heavy crude oil to more valuable, lighter products in refineries which are then left with petroleum coke (petcoke) as low-value by-product containing all the vanadium and nickel and about half of the sulfur in the original crude oil. World production of petcoke is rapidly increasing and expected to double every 10 years. Also its content of sulfur and heavy metals is expected to increase.

An increasing fraction of the petcoke and the heavy residual oil fractions must be used as fuel in the production of heat and power. This can most advantageously take place in the steam and power generation sections of the refineries or in nearby power plants.

All traditional flue gas desulfurization (FGD) technologies have the disadvantage of being increasingly more expensive to operate the higher the SO₂ content in the flue gas. In addition, the SO₃ and the metal oxide dust created by burning of petcoke give severe environmental and operational problems in traditional FGD plants.

Topsoe has developed a FGD technology, SNOX™ that has the opposite economical dependency on the SO₂ content in the flue gas: the more SO₂, the more economical the SNOX™ plant will be. The SNOX™ technology also solves the problems with SO₃ and metal oxide dust, and eliminates the emission of heavy metals. In a SNOX™ plant, the SO₂ in the flue gas is converted to concentrated sulfuric acid of high purity. This means that in a SNOX™ plant there is no consumption of absorbents, no production of waste materials that have to be deposited, and a product with a significant positive sales value. Furthermore, the SNOX™ process increases the overall steam and power production per unit of fuel.

The SNOX™ Process

The SNOX™ process is a regenerative, catalytic flue gas cleaning process which removes 95-99% of SO₂ and SO₃ and 90-96% of the NO_x in flue gases. The sulfur is recovered as 94-96% concentrated sulfuric acid of high purity. NO_x is catalytically reduced to N₂ by NH₃ added to the flue gas. Essentially all dust and particulates are removed from the flue gas. The heat produced in the process and by cooling of the flue gas to 100°C is recovered as steam and preheating of combustion air, thus increasing boiler thermal efficiency and gross power production.

The process generates no secondary sources of pollution such as waste water, slurries or solids. It consumes no water or materials, except for ammonia for the catalytic NO_x reduction.

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The process is in particular suited for purification of flue gas from combustion of high sulfur petroleum coke (petcoke) and other petroleum residues such as heavy fuel oil, tars and sour gases. In principle, there is no upper limit to the content of SO₂ and SO₃ in the flue gas.

The principal SNOX™ process steps are (see Fig. 1):

- Dust removal in ESP or bag filter at about 200°C
- Heating of the flue gas to about 400°C
- Catalytic reduction of NO_x by NH₃ added to the gas upstream of the NO_x reactor
- Catalytic oxidation of SO₂ to SO₃ in the subsequent oxidation reactor
- Further cooling of the gas to about 100°C in the WSA condenser, whereby the sulfuric acid vapor condenses

The process makes it possible to burn petroleum coke in PC (powdered coal) boilers without blending it with more coal or co-firing with more fuel oil than necessary for efficient combustion of the petcoke. Essentially all SO₃ and heavy metal oxides are removed from the flue gas without corrosion problems.

The process is especially suited for treatment of flue gas from burning up to 100% petcoke in downshot fired PC boilers. The high combustion temperatures and 3-4% excess O₂ required for high burn-out of petcoke and the high content of vanadium in the fly ash give high flue gas content of NO_x and SO₃, which both are efficiently removed without corrosion or other problems in a SNOX™ plant. The higher air preheat temperature with SNOX™ also improves burnout of the petcoke.

The main attraction of the SNOX™ process is that it makes it possible to burn cheap high sulfur petcoke for generation of steam and power without formation of waste products and with no more emission and operating problems than by burning normal coal with traditional FGD processes.

An added advantage is that, even before credit is taken for sales of the produced sulfuric acid, the operating cost of SNOX™ units slightly decreases with increasing SO_x content in the flue gas. This is due to the recovery of the heat of formation of H₂SO₄ from SO₂ amounting to 8 MJ/kg S in the fuel.

The first full scale SNOX™ plant treating 1,000,000 Nm³/h flue gas from a 300 MW coal fired power plant was started up in 1991 at Nordjyllandsværket in Denmark. In USA, SNOX™ was demonstrated in the scale of 35 MW on a coal fired power plant in Niles, Ohio, in 1991-96 under the DOE Clean Coal II program. Today, six SNOX™ plants and 55 WSA plants (i.e. SNOX™ plants without the DeNO_x step) treating a wide range of sulfur containing off-gases have been implemented worldwide by Topsøe.

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The largest SNOX™ plant treats up to 1,200,000 Nm³/h flue gas from four petcoke fired downshot boilers, built in the 1960s at the Agip Petroli refinery in Gela, Sicily, Italy. The SNOX™ plant was supplied on turnkey basis by Snamprogetti SpA and went into operation in September 1999. Topsoe supplied process engineering, catalysts and proprietary equipment. Originally the plant was designed for and operated with only three boilers, but in 2006, a 4th boiler was connected to the SNOX™ plant without any change of the plant.

The Experience with the SNOX™ Plant in Gela

The Gela plant lay-out is shown in Fig. 1.

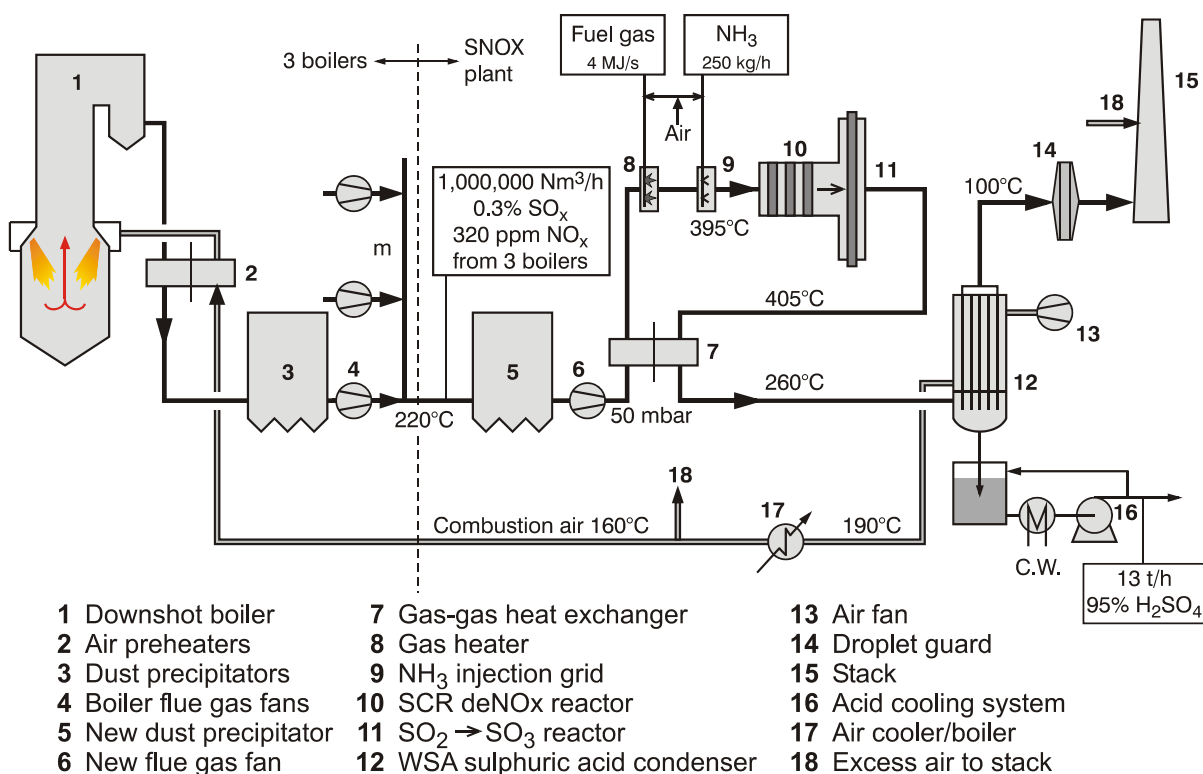


Fig. 1: The SNOX™ plant at the Gela Refinery treating flue gas from three utility boilers.

The SNOX™ plant, as seen to the right of the dotted line, treats the flue gas from three (later four) utility downshot boilers fueled with 1900 t/d of high-sulfur petroleum coke and minor amounts of fuel oil and refinery sour gases. Each boiler emits up to 370,000 Nm³/h flue gas and comprises an electrostatic precipitator (3) and a flue gas fan (4) which were retained when the boilers were retrofitted with the SNOX™ plant. The flue gas from the boilers is collected in the manifold (m) and passed to the new additional ESP (5).

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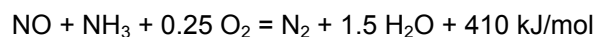
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The content of dust (mainly unburned petcoke) in the flue gas from the "old" ESP's is further reduced in the additional ESP to less than 1 mg/Nm³ of which 70% is carbon. The rest is ash of metal oxides (mainly vanadium oxide) most of which accumulate in the catalyst panels of the SO₂ reactor as oxy-sulfates.

After passing through the flue gas fan (6), the flue gas is heated in the rotating gas-gas heat exchanger (7) to 385°C and further to 395-400°C in the heater (8) using fuel gas (natural gas, refinery gases and/or H₂S gas from the refinery).

NOx Reduction

NH₃ (pre-mixed with hot air) is then injected and mixed with the flue gas through a grid of nozzles (9) placed in the flue gas duct upstream of the DeNOx reactor (10). In the DeNOx reactor, the gas flows horizontally through a vertical bed of monolithic DeNOx catalyst. Almost all of the NOx is present as NO which is reduced by the SCR reaction:

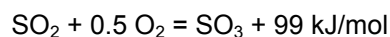


The degree of NOx removal obtained depends on the NH₃/NOx ratio and on how well the NH₃ is mixed with the flue gas. With typically 320 ppm NOx in the flue gas and a NH₃/NOx ratio of 1.00, the flue gas exiting the DeNOx reactor contains about 10 ppm NOx and 20 ppm NH₃. The NH₃ slip is destroyed in the SO₂ reactor.

SO₂ Oxidation

In the subsequent SO₂ oxidation reactor (11), the gas passes in parallel through 24 catalyst panels loaded with SO₂ oxidation catalyst in the form of "daisy"-shaped rings which give low pressure drop and high capacity for dust uptake. The DeNOx reactor and the SO₂ reactor are integrated in one single vessel.

In the SO₂ reactor, about 98% of the SO₂ is oxidized to SO₃:



At the same time, the NH₃ slip from the DeNOx reactor is completely oxidized to N₂ and NOx whereby 95% overall NOx removal measured after the SO₂ reactor is achieved. Higher hydrocarbons in the flue gas are also oxidized completely.

After 7 years of operation at 390-400°C, neither the DeNOx catalyst nor the SO₂ oxidation catalyst have deactivated significantly. Catalyst life time of up to 10 years is expected.

Final Dust Removal

At operating conditions, the sulfuric acid catalyst is slightly sticky and acts as an effective dust filter removing practically all of the remaining dust content. The catalyst is sticky because of the liquid pyrosulfate melt in the catalyst pores at operating conditions. The dust accumulates in the voids of the catalyst bed so that the catalyst in the panels must be cleaned at intervals that are inversely proportional to the dust content in the gas. The cleaning can be performed by circulating and screening the catalyst in a closed system, without interrupting the operation of the SNOX™ plant.

The ESP performs much better than expected and $< 1 \text{ mg/Nm}^3$ dust is found in the gas entering the SO₂ reactor. About 70% of the dust is carbon (unburned petcoke), which is oxidized to CO₂ when it gets in contact with the hot catalyst, so that $< 0.3 \text{ mg/Nm}^3$ of ash (mostly V₂O₅) from the gas phase accumulates in the catalyst beds. Hence, the catalyst is expected to operate 50,000 hours or more on full load between catalyst screenings. Actually, after 7 years of operation at average 90% load, less than 10-20% increase in pressure drop across the SO₂ reactor has been observed without the catalyst having been touched. The catalyst still looks clean with no visible dust, and it is expected that no catalyst cleaning will be necessary for another 2-4 years.

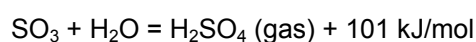
When heavy fuel oil is used as fuel, the dust load will be even lower. On the other hand, with normal coal having much higher ash content, more frequent catalyst screenings and use of the screening system is necessary, as in the Danish coal fired, SNOX™-equipped power plant.

Any trace of dust passing through the SO₂ reactor is removed by the condensing sulfuric acid in the WSA condenser and will end up in the product acid. The extremely low concentration of vanadium and the water-clear appearance of the acid confirm that no dust passes through the SO₂ reactor.

The WSA Condenser

After the SO₂ reactor, the gas is cooled to about 260°C in the rotating heat exchanger (7). The heat exchanger is equipped with recycle of sealing gas reducing the gas leakage to 2-2.5%. The leakage means that 98-98.5% SO₂ conversion obtained in the reactor decreases to 95.5-96% measured downstream of the heat exchanger.

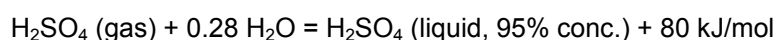
During the cooling, most of the SO₃ reacts with H₂O in the flue gas, forming H₂SO₄ vapor:



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The gas is well above the acid dew point when it enters the WSA condenser (12) which, in principle, is a falling film condenser in which the gas is further cooled to about 100°C in air cooled glass tubes. In the glass tubes, the remaining SO₃ is hydrated, and the sulfuric acid vapor condensed and concentrated to 95% strength. The mass and heat balance of the condensation is:



The heat of formation of sulfuric acid from SO₂ will increase the heat recovery by 0.5 MJ per kg of fuel if the fuel contains 6% sulfur.

The acid is collected in the bottom part of the condenser, from where it flows to the acid cooling system (16) where the acid is cooled to about 30°C in a water cooled plate heat exchanger.

The WSA condenser contains a large number of vertical glass tubes divided into modules. The cooling air is delivered by the air fan (13) and heated to about 190°C in the condenser. It is cooled in the boiler/trim cooler (17) to 150-160°C before it is heated in the air preheaters (2) and used as combustion air in the boilers. Excess air is passed to the stack. The boiler (17) is used to control the temperature of the flue gas entering the SNOX™ plant at about 200°C.

Any acid droplets in the stack gas are removed in a guard demister (14) installed in the duct upstream the stack.

Formation of acid mist (aerosol) in the condenser is avoided by heterogeneous nucleation control. The mist control is patented by Topsoe and is essential for the operation of WSA and SNOX™ plants.

The SNOX™-treated stack gas is invisible even against a blue sky, as seen in Fig 2.

Operating and Maintenance Experience

On the average, the SNOX™ plant has operated with 98% availability including planned stops, and no decrease in performance from the first start-up in September 1999 until now (January 2007) has been observed when measured conversions and pressure drops are corrected for changes in flow rate.

The results of the performance test run in February 2000 are summarized in Table 1.

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Inlet flue gas flow	971,000 Nm ³ /h
SO ₂ inlet	2885 ppm (molar)
NOx inlet	337 ppm (molar)
SOx removal efficiency	96.5 %
NOx removal efficiency	90.5 % ¹⁾
Acid mist in stack gas (prior to air dilution)	< 3 mol ppm "SO ₃ " ²⁾
Product acid concentration	95 % H ₂ SO ₄
Ammonia consumption	238 kg/h
Support fuel consumption (natural gas)	328 kg/h
Total power consumption (blowers, pumps, etc)	10.4 MW
Flue gas pressure drop across the SNOX™ plant	45 mbar
Stack gas opacity	< 5% (stack gas is invisible)
Product acid quality, ppm (recent measurements)	
Fe	2
Hg	< 0.005
Ni	0.065
V	0.035
Cr	< 0.03
As	0.09
SO ₂	< 10
HCl	< 10

¹⁾ NOx removal was later increased to 93-95% after injection grid adjustments.

²⁾ More accurate measurements later showed 1-2 ppm SO₃.

Table 1. Results of performance test run at the Gela SNOX™ plant.

The SNOX™ plant has been shut down for about a week every year for inspection and maintenance. The catalysts have not been touched, apart from topping up the panels with a small volume of catalyst to compensate for catalyst settling in the panels. In the summer of 2006, half of the catalyst volume was replaced with new catalyst, not because its performance was bad, but in order to secure another 3-5 years of trouble-free operation. The plant was always found to be in excellent shape and the catalysts with no signs of dust accumulation. No deformation or corrosion have been found in the reactors and the connecting ducts. The expansion joints were all in good shape.

Only a few glass tubes in the WSA condenser have broken during initial operation. A small number of broken glass tubes is of no consequence for the performance of the plant because the WSA condenser operates with higher pressure on the air side than on the flue gas side. A few signs of corrosion have been found and repaired, and later inspections have shown that the repairs are in good condition.

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The internal stack brick lining has remained in perfect shape and dry, confirming that essentially all acid droplets are removed before the flue gas enters the stack.

A weak spot in the design is the rotating gas-gas heat exchanger (7 in Fig. 1). Its sealing air system is often out of order, and then unconverted SO₂ gas is leaking into the converted gas, meaning a significant drop in overall SO₂ conversion efficiency. However, despite this internal leakage, the plant still fulfils the emission limits.



Fig. 2: The SNOX™ plant treating the flue gas from 3 boilers burning high-sulfur petroleum coke at the ENI refinery in Gela, Sicily, Italy. The stack to the left emits 1,000,000 Nm³/h of SNOX™ treated flue gas. The stack to the right emits flue gas from the 4th boiler before it was connected to the SNOX™ plant.

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New SNOX™ project

A new SNOX™ plant treating 820,000 Nm³/h flue gas from combustion of heavy residue in the power plant of the OMV refinery in Schwechat, Austria is under construction and scheduled for start up in the second half of 2007. The plant is designed with by-pass free gas-gas heat exchanger to achieve 98% SO_x removal.

Comparison with Alternative Processes

In practice petcoke cannot be used in ordinary PC boilers equipped with FGD scrubbers unless “diluted” with normal coal in the ratio 1 part petcoke to 4 parts coal based on heating value. Use of up to 100% high sulfur petcoke as fuel is only possible either in circulating fluid bed (CFB) boilers with injection of large excess of limestone in the combustion zone, or in downshot PC boilers. Gasification of petcoke in integrated gasification plants (IGCC) plants seems too expensive for power production.

CFB boilers is the prevailing technology for combustion of up to 100% petcoke for power production in spite of its disadvantages: Very high consumption of limestone and production of a correspondingly large amount of waste contaminated with all the vanadium and nickel from the petcoke, and the necessity of subsequent FGD (flue gas desulfurization) of the off- gas to achieve 98% SO₂ removal with high-sulfur fuels. To put it in perspective: 1 ton of fuel with 6% sulfur requires injection of 400-500 kg CaCO₃ and produces 600-700 kg heavy metal contaminated solid waste per ton of fuel. Use of SCR also seems necessary in order to achieve less than 40 ppm NO_x in the stack gas. If CFB boilers could operate without SO_x absorbent, the SO_x rich flue gas would be very suitable for downstream SNOX™ treatment.

Downshot boilers are excellent for combustion of petcoke. They can be designed for up to 600 MWe based on proven technology. However, the use of wet scrubbing technologies for treatment of the flue gas from combustion of high sulfur petcoke in PC boilers is hindered by serious problems (caused by the high content of SO₃ and the vanadium rich fly ash) with corrosion, plugging and mist formation which requires WESP (wet gas electrostatic precipitator) upstream the stack. Furthermore, usual “high dust” SCR NO_x reduction is hampered due to rapid fouling of the DeNO_x catalyst by the high vanadium content in the fly ash. The high consumption of limestone and other costs of achieving 98% SO_x removal is also a significant burden when scrubbing flue gas with high SO₂ concentration. Scrubbing technology has only been implemented in one case 18 years ago on a petcoke fired downshot boiler.

None of these problems are encountered with SNOX™ flue gas treatment which works in excellent synergy with petcoke fired in downshot boilers.

The SNOX™ lay-out with new downshot boilers will be similar to that shown in Fig. 1 with the modifications mentioned earlier, and no catalyst cleaning system will be necessary. Up to 99% sulfur removal and 96% NO_x removal can be achieved by using a leak-free (recuperative) gas-gas heat exchanger. The total investment cost per unit of net power production of a SNOX™-

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equipped downshot boiler will be approximately the same as that of a CFB boiler with the same SO₂ and NO_x emission, while the operating costs of the SNOX™-equipped boiler will, as seen in Table 2, be substantially lower, even before taking credit for the sales of sulfuric acid.

	Unit	Downshot PC boiler with SNOX™	CFB boiler with SCR and tail-end FGD
Net electric output	MW	300	300
Net electric efficiency (on LHV basis)	%	39	38
Petcoke burn-out efficiency	%	98	99
Petcoke consumption	t/h	88.3	89.7
SO _x removal efficiency	%	98	98
NO _x content in stack gas	ppm	< 20	30
H ₂ SO ₄ production	t/h	15.6	0
Total Ca/S ratio in SO _x absorption		-	2.8
CaCO ₃ consumption	t/h	0	47 ¹⁾
Waste production	t/h	0	75 ²⁾
NH ₃ consumption	t/h	0.25	0.1
Water consumption	m ³ /h	0	?
Waste water production	m ³ /h	0	?
Operating costs in USD per year at 8000 full load hours per year:			
H ₂ SO ₄ sold at 15 \$/t	\$/y	-1,800,000 ⁴⁾	0
CaCO ₃ supplied at 20 \$/t	\$/y	0	7,500,000
NH ₃ supplied at 300 \$/t	\$/y	600,000	240,000
Water consumption	\$/y	0	?
Waste water disposal	\$/y	0	?
Waste disposal cost at 20 \$/t ³⁾	\$/y	0	12,000,000
Cost of difference in fuel consumption at 40 \$/t petcoke	\$/y	0	400,000
Net cost of materials per year	\$/y	-1,200,000⁴⁾	~ 20,000,000
Net cost of materials per MWh	\$/MWh	-0.5⁴⁾	8.3

¹⁾ Use of CaO containing CFB waste for tail gas FGD is assumed

²⁾ Mixture of CaSO₄, CaO, CaCO₃ and ash. The weight depends on the H₂O-content

³⁾ Typical figure that varies considerably with the location

⁴⁾ Negative figures mean positive income

Table 2. Comparison of a SNOX™-equipped downshot boiler with a CFB boiler with SCR DeNO_x and tail-end FGD for producing net 300 MW electricity from petcoke with 6 % sulfur and an LHV of 32 MJ/kg.

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Conclusions

After more than 7 years of operation, the SNOX™ plant treating the flue gas from 4 petcoke fired downshot boilers at the Agip Petroli refinery in Gela has demonstrated high SO_x and NO_x removal from the flue gas from combustion of high sulfur petcoke. There are no significant corrosion problems, no problems with corrosion or fouling and there has been no decrease in the performance of the SNOX™ plant. The treated stack gas is invisible and essentially free of SO₃ and heavy metals. SO₂ and SO₃ are recovered as commercial grade 95% H₂SO₄ of high purity. The heavy metals (vanadium and nickel) are separated as a dry concentrate of oxides. The availability of the SNOX™ plant including planned outages has been 98% since the start. Thus, it can be said that SNOX™-equipped downshot fired boilers are a technically and environmentally superior solution for power plants burning high sulfur petcoke. Formation of waste products is avoided, and no more emissions occur than with burning ordinary coal in usual FGD-equipped boilers.

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