THE APPLICATION OF A LOW TEMPERATURE SELECTIVE CATALYTIC REDUCTION SYSTEM FOR MUNICIPAL & HAZARDOUS WASTE COMBUSTORS

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ABSTRACT

In Central Europe during the late 1980's and through the early 1990's, emission regulations on municipal and hazardous waste combustors (MWCs and HWCs) were tightened drastically. Among other pollutants, NO_x emissions had to be limited to an extent that required the installation of special NO_x control technologies and 70 mg NO_x/Nm^3 (56 ppmdv) (corrected to 11% O_2 if the measured value exceeded 11% O_2). This became a commonly accepted value for most permitting agencies in Germany, Holland, Austria and Switzerland. The Selective Catalytic Reduction (SCR) technology became the preferred NO_x control technology for retrofitting existing MWCs and HWCs, as well as for new facilities.

This paper presents the Low Temperature SCR technology (LTSCR) as a major new development in SCR technology adapted to MWCs and HWCs. LTSCR's can be operated at temperatures as low as 150°C (302°F) while SCR's operate at temperatures above 280°C (536°F). The paper outlines the specific needs and restrictions of LTSCR, as well as its advantages. A detailed description of the correlation between required volume of catalyst, temperature, and specific catalytic activity is given. The application of LTSCR is shown for MWCs and HWCs, and for each case, one retrofit and one new facility are introduced. Finally, the paper reports on some two and a half years of operating experience with LTSCR and gives an outlook on further applications.

INTRODUCTION

The emission of NO_x is currently by far the largest single air pollutant contributing to the acid rain problems in Germany since FGD systems were installed on all coal-fired utility boilers above 50 MW (thermal). In order to reduce NO_x emissions in Germany, several legislative steps were taken during the 1980's and early 1990's. The emissions from power plants and other large sources were limited to such an extent that almost all facilities had to be equipped with low-NO_x burners, SCR-DeNO$_x$ systems, or other NO_x reduction technologies.

For municipal and hazardous waste combustors (MWCs and HWCs) emission limits were also tightened drastically. Table 1 gives an overview of German NO_x legislation for MWCs and HWCs between 1974 and the present, and a comparison with U.S. Environmental Protection Agency (EPA) emission limits.

As noted in Table 1, the NO_x emission guarantees required from vendors of air pollution control (APC) equipment are a factor of four or more below the German Federal Standard. This is a result of political intervention caused by public pressure on local permitting agencies by groups such as the Green Party followed by Regions requiring the Best Available Control Technology (BACT).

Known and well proven from power plant applications, SCR soon became the predominant choice for retrofitting existing MWCs and HWCs, as well as for new facilities. The first two MWCs retrofitted with SCR plants in Germany were the Munich-South plant and the Stuttgart-Münster plant. In both cases, the same circumstances led to choosing SCR as the only feasible option for NO_x reduction. These circumstances are summarized as:

- Both plants were located and operated together with a power plant equipped with SCR;

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- For both plants, the NO$_x$ emission limits were set at 70 mg/Nm^3 (56 ppmdv) by local requirements [4];
- Both plants were owned and operated by its city's utility company;
- In both plants, the installed boilers were not suitable for a Selective Non-Catalytic Reduction (SNCR) DeNO$_x$ system due to the boiler design and low combustion temperature in older designs.

Based on these facts, the most logical choice was to add a Low-Dust SCR plant at the tail end of the existing APC-train. Low Dust SCR systems are usually defined as systems that are installed after the primary electrostatic precipitator (ESP) and the FGD plant while the High-Dust SCR system operates before the economizer of the boiler and the ESP. The SCR plants in both MWC facilities then resembled almost exactly the well known, well proven, Low-Dust SCRs in power plants.

Figure 1 shows the typical arrangement of a Low-Dust SCR plant operating at approximately 300°C (572°F) and employing a regenerative type rotary heat exchanger (Ljungström System) [5]. Here the flue gas coming from the APC-train is heated to approximately 280°C (536°F) by the 300°C (572°F) hot flue gas leaving the SCR. In order to overcome the gradient of the heat exchanger, the final heating from 280°C (536°F) to 300°C (572°F) is done by a natural gas burner. Then gaseous ammonia or anhydrous ammonia (NH$_3$ or NH$_4$OH) is injected into the flue gas prior to the catalysts for the reduction of NO$_x$ as detailed later in this paper. These SCR plants have been in operation for more than 30,000 hours and have proven to be highly effective, low maintenance, and easy to operate systems. However, significant cost for investment as well as operation are entailed. Due to these disadvantages, it became desirable to develop an SCR technology aimed towards lower costs while still maintaining its advantages.

SCR LIMITATIONS

The major disadvantage of the application of SCR is the high operating temperature which is set by the maximum conversion efficiency. Figure 2 shows that the optimum temperature ranges between 300°C (572°F) and 410°C (770°F). However, operation above 350°C (662°F) is not recommended due to increased brittleness of the catalyst and softening of the ash. Below and above that temperature, a sharp decrease in conversion efficiency occurs. To be avoided, this decrease has to be compensated for by a higher volume of catalyst or a higher reactivity of the catalyst. Associated with higher catalyst volumes is an increase in investment for the catalyst and for the SCR reactor. This increase is partially offset by a decrease in required heat exchanger surface area due to a smaller amount of thermal energy to be transferred. A higher pressure drop caused by more catalyst employed is almost fully compensated by a lower pressure drop across a smaller heat exchanger. As shown in Figure 3, the average life expectancy of the catalyst is a major overall cost influencing factor. The lifetime of the catalyst,

however, depends on the purity of the flue gas entering the catalyst and on the operational temperature due to physical/dimensional reasons.

The catalytic reactivity of any catalyst is susceptible to catalytic poisoning. It is caused by certain substances which permanently attach themselves to the catalytic surface, thus blocking their catalytic reactivity. Some heavy metals such as arsenic, as well as alkaline elements such as sodium and potassium, are known to cause a permanent irreversible disactivation of SCR catalysts. Therefore, it is advantageous for the lifetime of the catalyst to preclean the flue gas as much as possible.

A poisoning of the SCR catalyst that can be reversed is caused by a deposition of ammonium salts such as ammonium chloride (NH$_4$Cl) and ammonium sulfate ((NH$_4$)$_2$SO$_4$). These salts are formed from the residual hydrogen chloride (HCl) and sulfur trioxide (SO$_3$) still present in the flue gas after the APC system and the injection of ammonia (NH$_3$). Additional ammonium sulfate ((NH$_4$)$_2$SO$_4$) is formed after partial oxidation of sulfur dioxide (SO$_2$) to sulfur trioxide (SO$_3$) by the catalyst. This process is commonly referred to as SO$_2$/SO$_3$ conversion. The degree of SO$_2$/SO$_3$ conversion also increases with the increasing reactivity of the catalyst which creates a problem addressed later in this paper.

The ammonium salts have a tendency to condense and deposit on the catalytic surfaces at lower temperatures. Especially critical is the formation and deposition of ammonium hydrogen sulfate (NH$_4$HSO$_4$) which typically occurs at temperatures around and below 170°C (338°F), depending on its concentrations. It then forms a sticky deposit that clogs the porous surface of the catalyst. The process can be reversed by "cooking" the catalyst at temperatures above 300°C (572°F) by using auxiliary burners thus evaporating the salt. Since this regenerating procedure is costly and time consuming, the avoidance of the problem is the better solution. Figure 4 shows the formation characteristics of ammonium sulfate ((NH$_4$)$_2$SO$_4$) and ammonium hydrogen sulfate (NH$_4$HSO$_4$) depending on SO$_3$ and NH$_3$ concentrations at various temperatures [6]. As can be seen, the presence of SO$_2$/SO$_3$ has to be greatly reduced in order to avoid the salt formation and especially deposition at lower temperatures.

The SO$_3$ concentration does not only depend on the SO$_3$ removal efficiency of the preceding APC-train. It also depends on the still remaining SO$_2$ concentration and the SO$_2$/SO$_3$ conversion within the SCR reactor. As SO$_2$/SO$_3$ conversion increases with the increasing reactivity of the catalyst, this poses a problem with LTSCR. Due to the fact that the LTSCR catalyst has a much higher reactivity than a "normal" SCR catalyst (in order to counteract the required increase in catalyst volume), it also has a much higher SO$_2$/SO$_3$ conversion rate. However, since the higher reactivity is a prerequisite for LTSCR due to cost considerations, the SO$_2$ removal requirements for the preceding APC-trains are increased significantly.

In order to avoid the ammonium salt deposition problem in an LTSCR plant, the flue gas has to be virtually free of SO_2 and SO_3 prior to the LTSCR plant. Even though this restricts the applicability of LTSCR, the cost aspect clearly demands the increased reactivity of the LTSCR catalyst. Figure 5 provides a comparison between the LTSCR catalyst and a "normal" catalyst for a given NO_x reduction efficiency of $\eta = 0.8$ [5]. It clearly shows that lower operating temperatures are not feasible employing a "normal" catalyst since the volume increases exponentially. The LTSCR catalyst volume increase is significantly less drastic. This is achieved mainly by raising the concentration of the catalytically active compound vanadium pentoxide (V_2O_5) in the titanium dioxide (TiO_2) based catalyst from 2-3% in a "normal" SCR catalyst to 10% or more in LTSCR catalysts.

THE DEVELOPMENT OF LTSCR

With all the limitations and considerations outlined above, the obvious question arises, "Why bother lowering the operating temperature of the SCR plant at all"? The solutions to resolve the SCR limitations led to the development of LTSCR.

The Reheat Problem

As explained above, the flue gas coming from the APC-train needs to be reheated to the "normal" SCR temperature of approximately 300°C (572°F). Even though most of the heat transfer is managed by the heat exchanger to minimize heat losses, the gradient of the heat exchanger still has to be overcome. Therefore, an external heat source in the form of an natural gas and/or oil fired burner or a high pressure steam heater must be used. The steam parameters commonly found in MWC and HWC boilers range from approximately 280°C and 2 MPa (536°F + 290 psi) up to approximately 400°C and 4 MPa (752°F + 580 psi). This steam, in any case, is not suitable for heating flue gas to more than approximately 230°C (446°F). This means that a natural gas and/or oil fired burner has to be installed in order to achieve a flue gas temperature of more than 230°C (446°F). The use of such expensive fuels results in a significant contribution to the overall operating cost. Since low pressure steam produced in the heat recovery boiler of MWCs and HWCs is of relatively little value in generating electric power, it is desirable to operate the SCR at temperatures compatible with a facility's steam capabilities, usually 230°C (446°F) or lower.

The Flue Gas Polishing Requirement

For retrofitting many existing facilities, as well as for all new facilities, it became politically desirable in Germany as well as in Holland, Austria and Switzerland to reduce all air pollutants to the maximum possible extent. Applying BACT in order to reduce all emissions to values around the detection limited laid the ground for LTSCR. All restrictions and limitations concerning the poisoning of the catalysts disappeared to a large extent with the introduction of flue gas polishing stages in the

APC-train. As published earlier [7,8,9,10], this equipment, employing activated carbon based materials or a lime enhanced product such as Sorbalit, leads to flue gas virtually free of heavy metals, organics, and acid gases. With a flue gas composition similar to that from natural gas fired units, the breakthrough for LTSCR was achievable.

The Cost Criteria

In order to meet the extremely low emission levels required in Germany, as well as in Holland, Austria and Switzerland, expensive complex multi-stage APC systems are employed. These complex 3-4 stage APC systems reduce the particulate emissions and other contaminates to such low levels that they do not poison the catalyst. The initial high capital and operating cost of high efficiency NO_x control can be offset by the cost of the preceding APC-train and flue gas polishing stage followed by the LTSCR. These complex APC systems make the LTSCR technology economically and technically viable.

The capital cost of these systems are reduced when the operating temperature of the LTSCR is fairly close to that of the preceding polishing stage. The polishing stage is commonly operated around 130°C (266°F) to 140°C (284°F). With a feasible operating temperature of a LTSCR plant between 150°C (302°F) and 180°C (356°F) and a minimum gradient of any heat exchanger of 15-25K (degrees Kelvin) to be overcome by a steam heater, the installation of a heat exchanger becomes unnecessary. The reduced capital cost is due to the elimination of the heat exchanger and the lower operating temperature which allows for the use of carbon steel in the duct work and the LTSCR reactor, as well as the lower cost of the insulation. Finally, the high purity of the flue gas leads to a lifetime expectancy of the catalyst in excess of 10 years, compared to the lifetime expectancy in an SCR facility of 5 years, thus greatly reducing the operating cost.

Based on these considerations, two principally different types of SCR technologies developed. One is the LTSCR with typical operating temperatures between 160°C (320°F) and 175°C (347°F), in some special cases up to 200°C (392°F), and the Combination SCR with typical operating temperatures ranging from 280°C (536°F) to 320°C (608°F). The name Combination SCR results from the fact that the SCR reactor is designed to not only reduce NO_x but also to destroy organics such as dioxins and furans by means of catalytic oxidation. Figure 6 provides an overall cost comparison between LTSCR and a "normal" SCR. The economic break point is shown to be at 230°C (446°F) which is approximately the temperature where steam heating becomes more economical than natural gas/oil fired burners.

Figure 6 also indicates clearly why the majority of new MWCs and HWCs are equipped with LTSCR's and most retrofits on MWCs and HWCs are either of the LTSCR type or Combination SCR. Operating temperatures are either below 200°C (392°F) or above 280°C (536°F) for obvious reasons.

Typical LTSCR Applications for MWCs

Through the 1980's, a typical MWC plant was equipped with an APC-train suitable for achieving the emission values similar to the ones currently applied in the U.S. Therefore, dry or semi-dry systems were predominant. These systems usually consist of one of the following combinations:

- ESP/spray dryer/baghouse
- ESP/spray dryer/ESP
- Spray dryer/baghouse
- Spray dryer/ESP
- Dry injection/baghouse
- Quench cooler/dry injection/baghouse

Hence, these systems reached their limits requiring retrofits in order to meet the new legislation. A common way of retrofitting these APC-trains was to add on an Activated Char Reactor (ACR) and a LTSCR plant. Figure 7 shows such a system. The flue gas temperature at the outlet of the baghouse is 140°C (284°F) thus it is perfectly suited for the ACR. After the ACR, the ID fans add about 10K to the flue gas due to the heat of compression. In order to reach the LTSCR temperature of 160°C (320°F), a steam heater is employed. After passing through the SCR reactor, the flue gas is discharged directly through the stack which is located directly above the SCR reactor. Figure 8 gives an overview of this effective and cost saving arrangement. The main cost reducing factor is the combination of the LTSCR reactor and the stack.

An advantage to this system process is the flue gas temperature of about 140°C (284°F) coming from the spray dryer/baghouse. Therefore, no reheating of the flue gas is required prior to the ACR. Since new facilities utilize mostly wet scrubbers, a cross-flow heat exchanger employing Teflon Tubes is commonly used to raise the temperature to 120°C (248°) to 140°C (284°F). Figure 9 provides a process flow diagram of a five-stage complex APC-train discussed earlier [11]. Due to the relatively low temperature of 200°C (392°F) of the flue gas entering the cross-flow heat exchanger and the flue gas being fully saturated at about 65°C (149°F) exiting the second scrubber, the maximum achievable temperature prior to the ACR is around 120°C (248°F) to 130°C (266°F). However, with the significant pressure drop of such a system of well over 10 kPa, the ID fans after the ACR unit add at least 10K to the flue gas. With an operating temperature of 160°C (320°F) the steam heater has to supply only 20K to 30K to the flue gas utilizing low pressure steam. The plant arrangement as shown in Figure 10 is similar to the described retrofit as far as the LTSCR is concerned. The space saving, economical combination of the LTSCR reactor and the stack will be realized in this MWC facility as well.

Typical LTSCR Application for HWCs

In contrast to MWCs, HWCs generally have a much higher concentration of acid gases and other pollutants at the inlet of the APC-train. Hence, an ESP followed by multiple stage wet scrubbers are traditionally employed. Due to possible condensation of metal salts formed in the flue gas causing clogging of the ESP, these are commonly operated at temperatures above 230°C (446°F) to 240°C (464°F). For the same reason, cross-flow heat exchangers are not being used in HWCs before the scrubbers. This leads to the fact that the flue gas exiting the scrubbers commonly has a temperature of about 70°C (158°F).

In order to minimize heat losses an ACR/LTSCR arrangement involving one counter cross-flow heat exchanger is used. This arrangement is commonly used to retrofit existing HWCs as presented in Figure 11 as well as for new HWCs as outlined in Figure 12. The main difference is the location of the fan. In the retrofit case, the existing APC-train is equipped with one fan after the scrubbers. The retrofitted ACR/LTSCR plant utilizes a second ID fan prior to the stack. In the case of a new HWC facility, the more economical solution of only one fan is used for the entire train. In order to utilize the significant temperature increase across the fan, due to the high pressure increase across the fan, it is located between the ACR and the LTSCR plant.

In the retrofit case, the flue gas enters the heat exchanger fully saturated at around 70°C (158°F) and is heated to around 140°C (284°F). The steam heater after the ACR adjusts the flue gas to the LTSCR temperature of 170°C (338°F). After passing through the heat exchanger again, the flue gas reaches a final temperature of about 100°C (212°F) before the second fan. Since the pressure drop to be overcome by the second fan does not result in a significant heat gain, it is more economical to operate the fan at 100°C (212°F) than at 140°C (284°F) due to the smaller actual volume flow rate and, therefore, the smaller power consumption.

This system is tailored to minimize operating costs, which can only be done in the unspoiled situation of designing an APC-train from the ground up. The flue gas coming from the scrubber is heated to around 100°C (212°F) by a steam heater. It is operated as the second stage of the steam heater prior to the LTSCR reactor. After that the flue gas enters the heat exchanger to be heated to around 130°C (266°F). After the ACR unit, the fan adds approximately another 10K prior to the steam heater upstream of the LTSCR which brings the flue gas to its final temperature of 160°C (320°F).

This arrangement also requires less duct work than a new facility. Here the entire system around the LTSCR is optimized from the beginning. Figure 13 shows the general arrangement of that APC-train. As can be seen, locating the fan between the ACR and the LTSCR requires a little more duct work than in the retrofit case, but this is well paid for by the reduced steam consumption and the smaller counter cross-flow heat exchanger.

Operating Experience

Seven LTSCR systems in Germany and Holland have been in operation now for two to three years. Problems arose only in one case where a leakage through a bypass damper allowed SO_2

contaminated flue gas to enter the LTSCR [12]. The resulting deposition of ammonium hydrogen sulfate (NH_4HSO_4) on the catalysts operated around 150°C (302°F) caused a fast deactivation of the catalyst. By "cooking" the catalysts with gas burners at 300°C (572°F) for 24 hours, the full reactivity was restored. This incident clearly proved the suspected high susceptibility of LTSCR to SO_2/SO_3 in the flue gas especially at temperatures below 170°C (338°F).

Reactivity tests of LTSCR catalysts were only obtained during the first year of operation due to some uncertainty about the new technology. Since none of the reactivity tests showed any decrease in activity and operating performance remained unchanged, no further tests were conducted. Table 2 gives some results of reactivity testing of one of the first LTSCR catalysts operating on a full-scale basis. The start-up of this particular plant in Rotterdam, Holland was in 1993. The LTSCR has operated with unchanged performance ever since. By the end of 1993, there were more than 15 APC-trains with LTSCR operating in Germany, Holland and Austria, and more than 30 under construction and in the planning phase.

CONCLUSIONS

The LTSCR technology has proven to be an economical and technical alternative to NO_x control by SCR technology to meet the stringent emission limits currently being employed in Western Europe. The LTSCR has a lower capital and operating cost than conventional SCR applications when low emissions of particulates, SO_2, HCl, and organics is required.

The LTSCR technology may have applications in the U.S. under the rules promulgated by EPA on October 31, 1995 for MWCs. In these rules, EPA allowed for NO_x emission averaging or "the bubble concept" for facilities with two or more units. While most SNCR technologies generally achieve a 50% reduction, LTSCR technology reduces NO_x emissions in excess of 90%. In concept, this means that if an MWC facility has two units, only one unit would have to be retrofitted with an LTSCR while the other unit can remain without NO_x controls.

Another economic application to the U.S. market for the LTSCR technology will be the ability to sell NO_x credits. The application of SNCR in MWCs will not produce sufficient reduction to generate NO_x credits. EPA has also made provision for non-contiguous MWCs to trade NO_x allowance credits. The application of the LTSCR technology in MWCs with the greater reductions will provide significantly more tradeable credits than the SNCR technology.

Other applications in the U.S. of LTSCR could be at Boiler Industrial Furnaces (BIFs) and cement kilns that burn hazardous waste. For these applications, extremely low levels of pollutant emissions will be required in order to achieve political and public acceptance. As yet, the U.S. has not promulgated emissions limits for these facilities. However, for applications which require the lowest achievable emission rates, LTSCR has already proven itself to be a practical and economical alternative to reducing NO_x emissions.

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Experience" presented at the 85th Annual AWMA Meeting, Kansas City, MO, 1992.

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TABLE 1

GERMAN AND U.S. NO_x EMISSIONS REQUIREMENTS FOR MWCs and HWCs

Standard Set By	TAL '74 [1]	TAL '86 [2]	17th BImSchV [3]	Local Permitting Requirements [4]	German Vendor Guarantees	U.S. EPA MWC¹ [13]
Date	1974	1986	1990	1992	1995	1995
NO_x emission limit	none	500 (400)	200 (160)	70 (56)	30 - 50 (24 - 40)	262 (210)

All values are in mg/Nm^3 (ppmdv) and actual O_2 or corrected to 11% O_2 if the measured value exceeded 11% O_2

TABLE 2

REACTIVITY TEST RESULTS OF LTSCR CATALYSTS

APC-Train No.	Hours of Operation	NO_x Outlet (ppmv)	% NO_x Actual Reduction	% NO_x Design Reduction	K-factor (m/h)	Ko-factor (m/h)	K/Ko
1	5202	63	68.5%	65%	23.3	21.1	>1
2	5382	72	64%	65%	20.6	21.1	0.98
3	6079	66	67%	65%	22.4	21.1	>1

Tests on @ T=210°C (410°F)
NH_3/NO_x = 1
Inlet NO_x 200 ppmv
O_2 = 9.2% (dry basis)
H_2O = 15% (wet basis)
K-factor: space velocity actual factor m/h
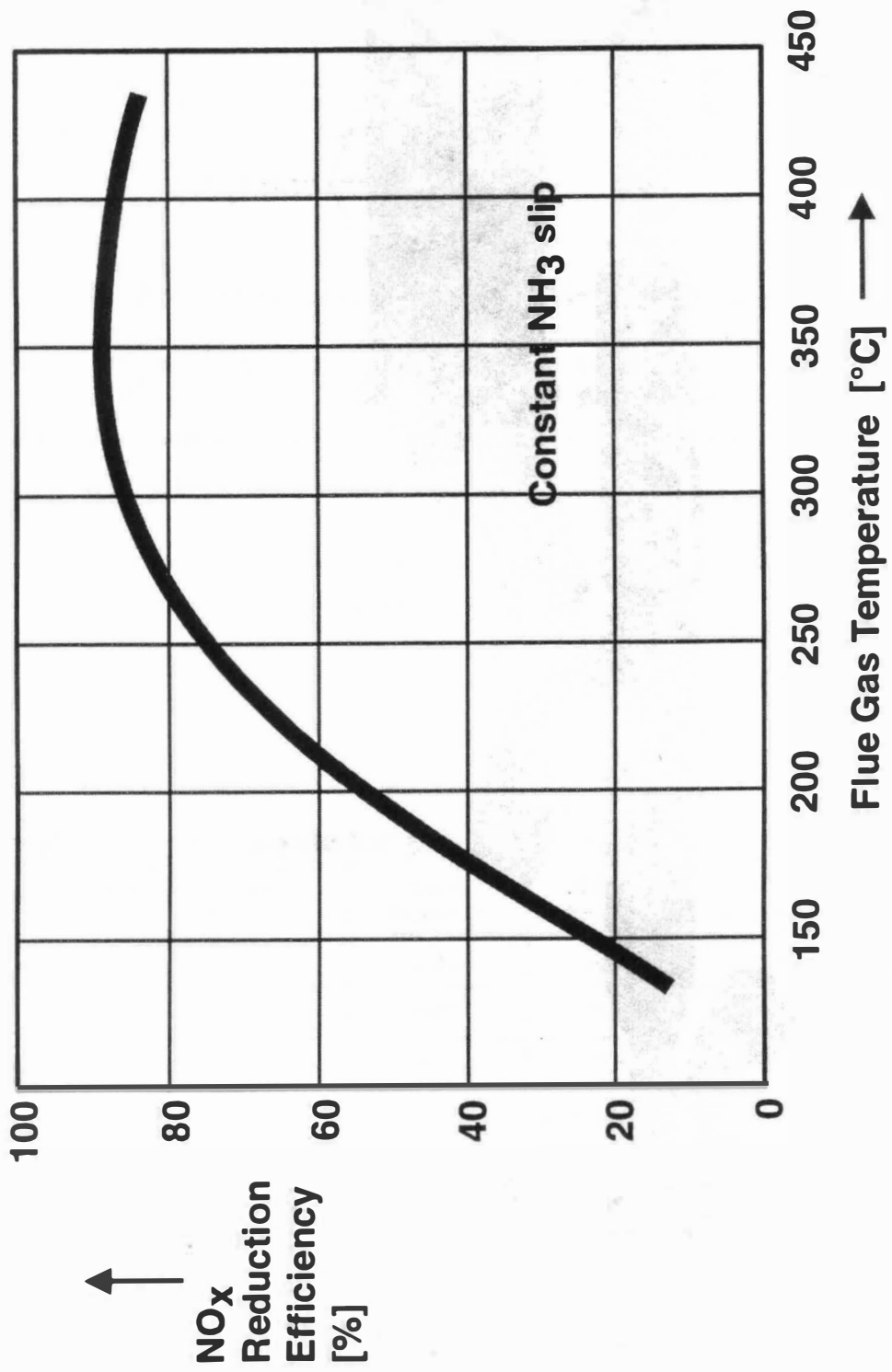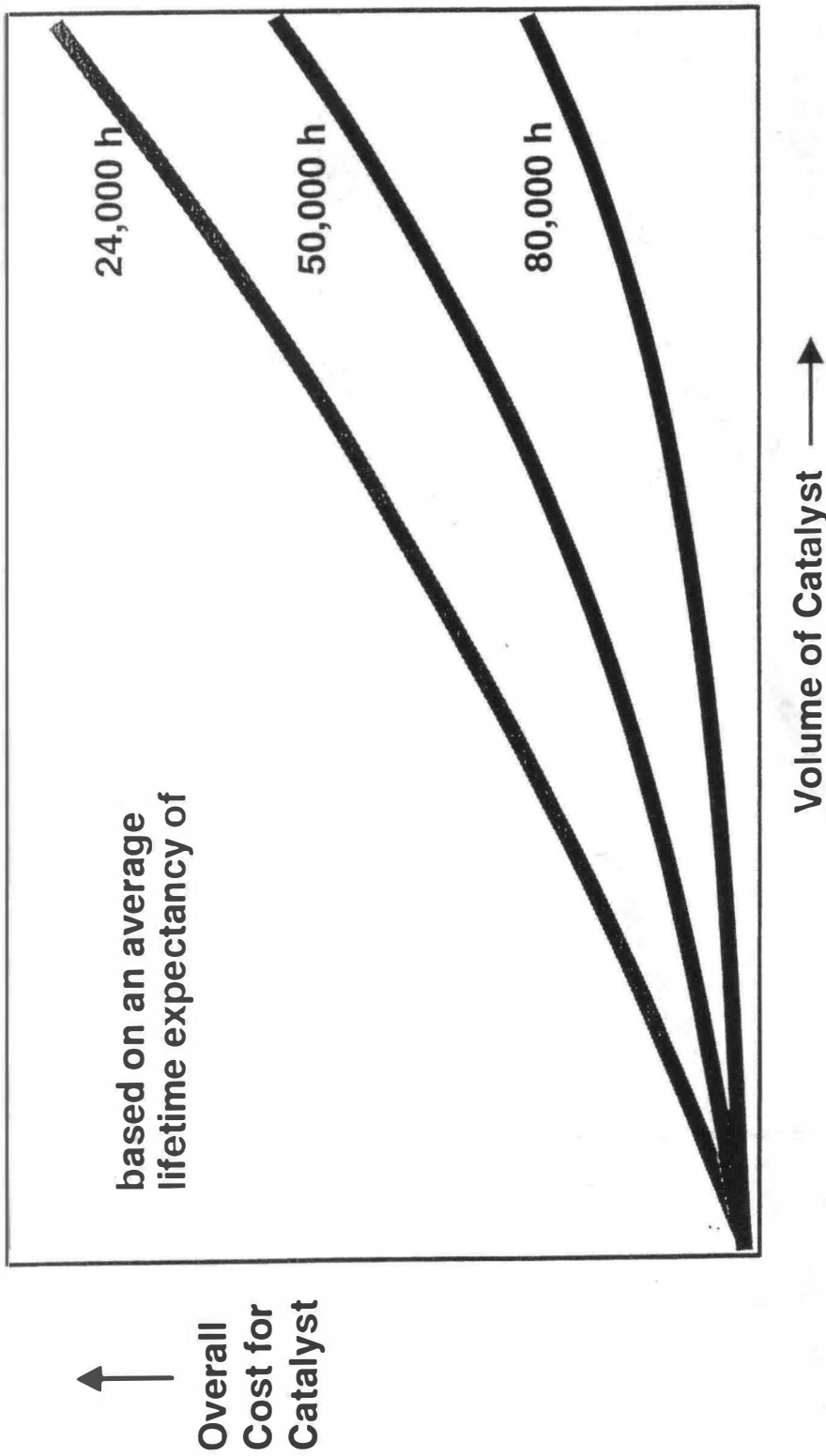Ko-factor: space velocity design factor m/h

NH₃ injection system

catalysts

steam heater or gas/oil burner

rotary heat exchanger

to stack

from APC-train

ID-fan
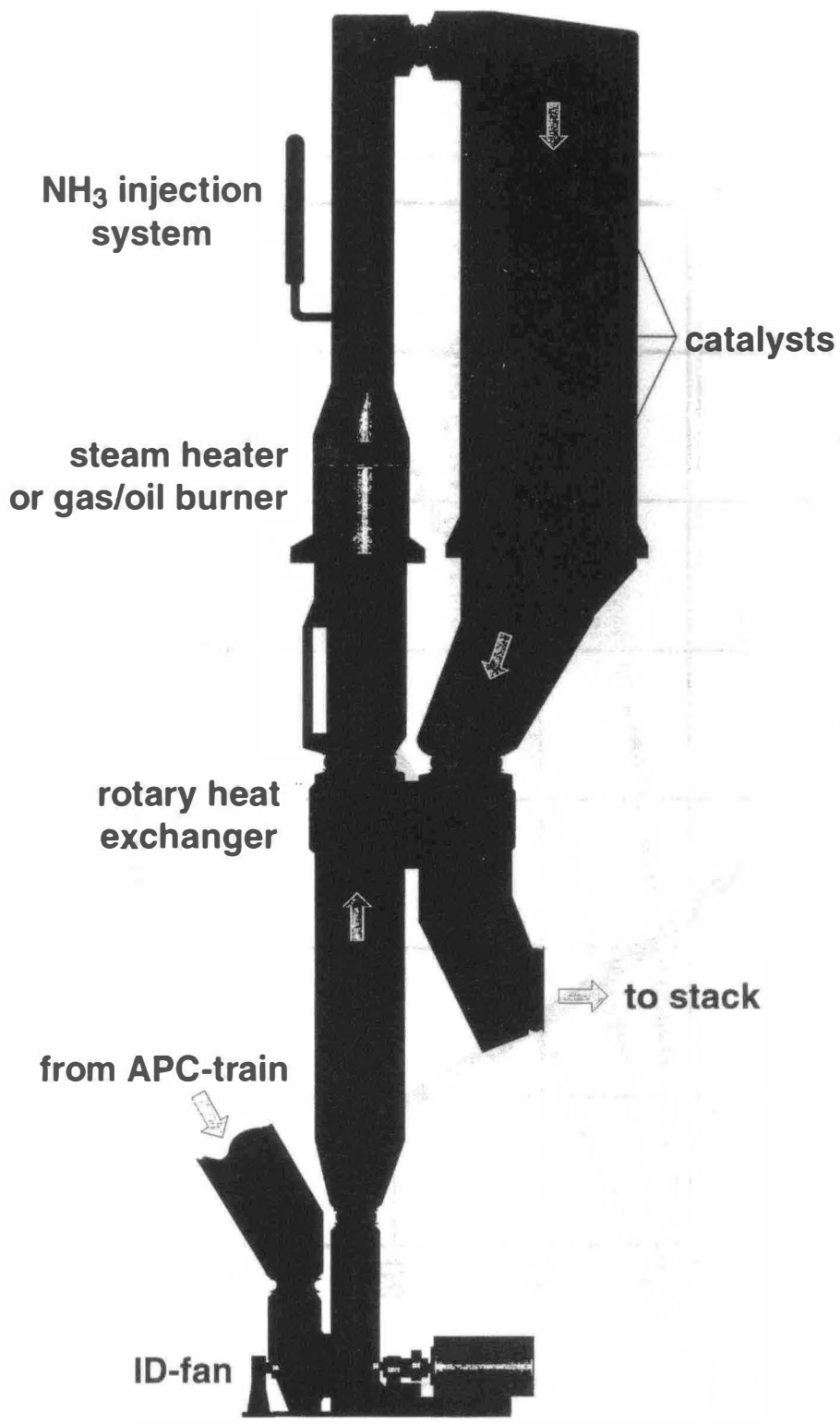
FIG. 1 Typical Low Dust SCR-Plant

FIG. 2 NO$_x$-Reduction Efficiency as a Function of Temperature

based on an average
lifetime expectancy of

24,000 h

50,000 h

80,000 h

Overall
Cost for
Catalyst

Volume of Catalyst ⟶

**FIG. 3 Influence of the Average Lifetime Expectancy
of Catalyst on the Overall Cost for Catalyst**

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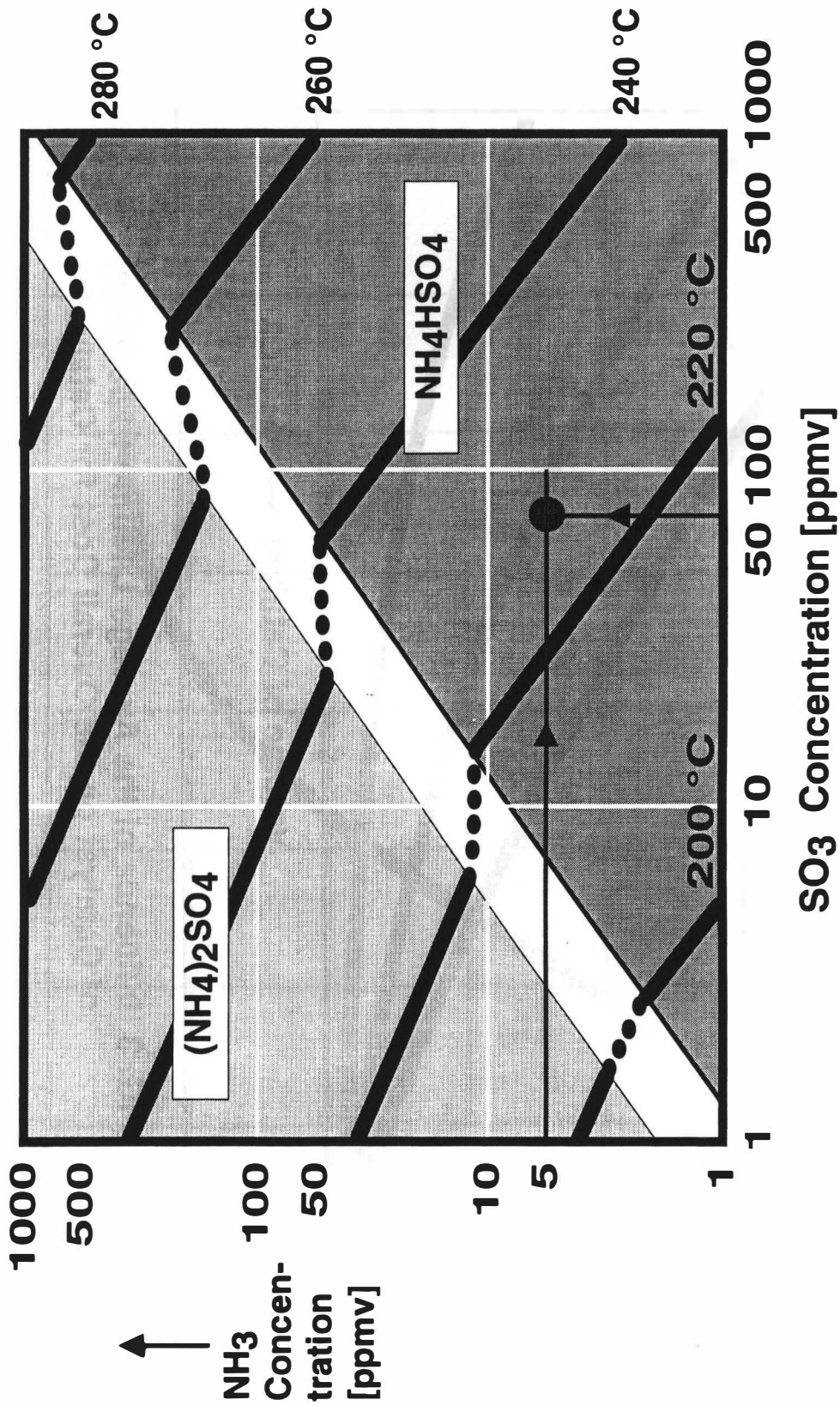
FIG. 4 Formation and Precipitation Characteristics of Ammonia Sulfate and Ammonia Hydrogen Sulfate

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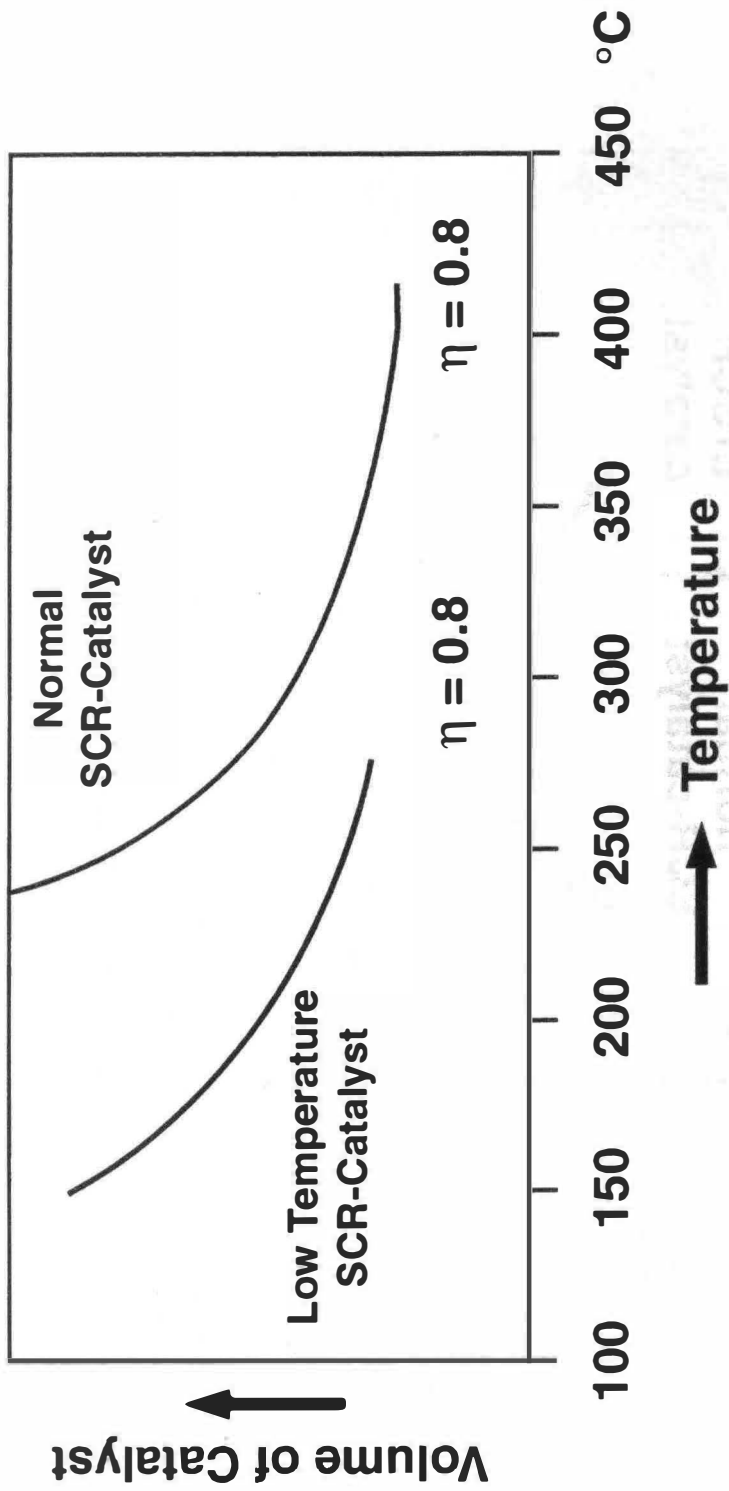
FIG. 5 Required Catalyst Volume Depending on the Flue Gas Temperature for Normal and LTSCR Catalysts
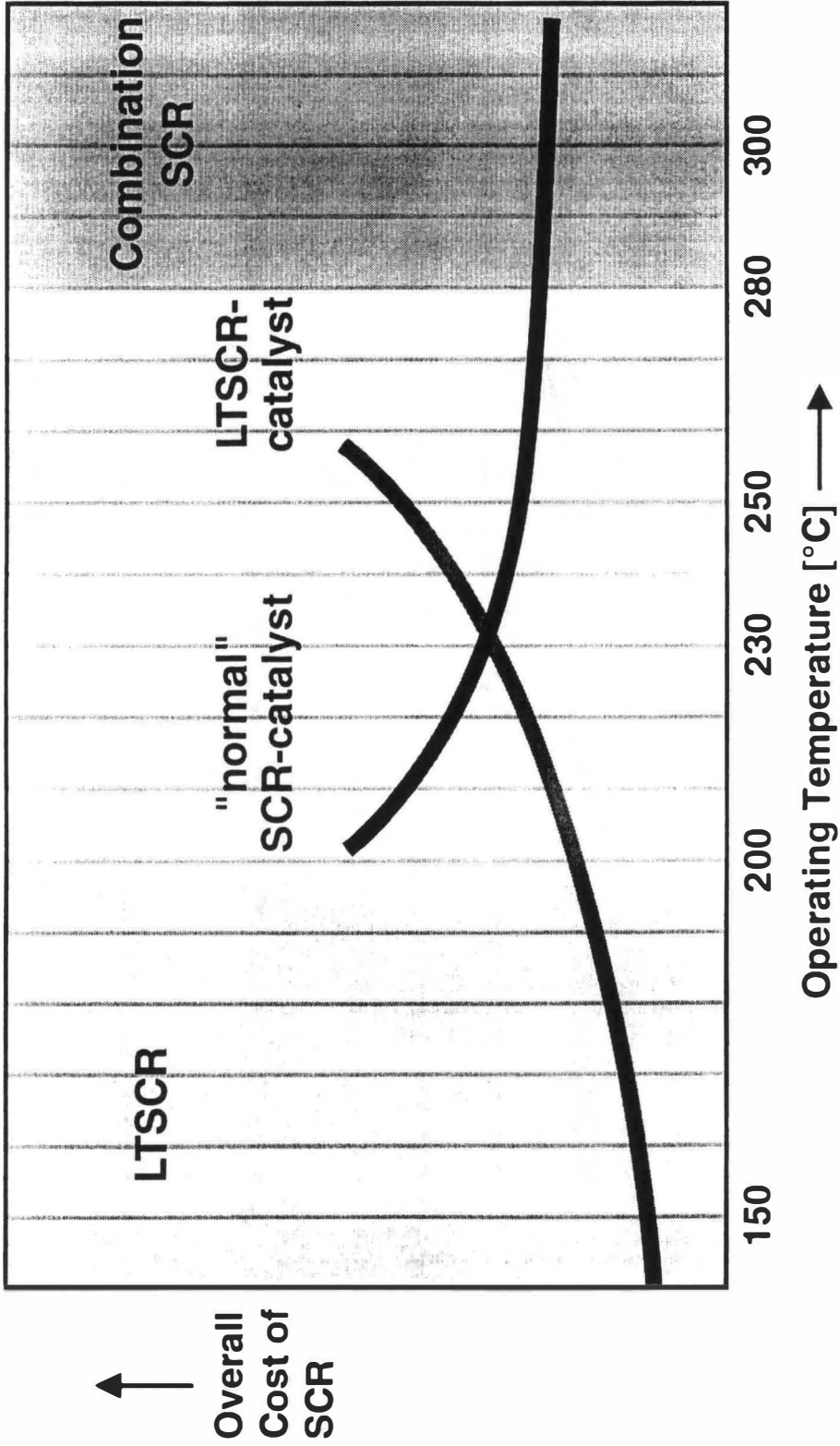
FIG. 6 Overall Cost Comparison of LTSCR and "Normal" SCR Depending on the Operating Temperature
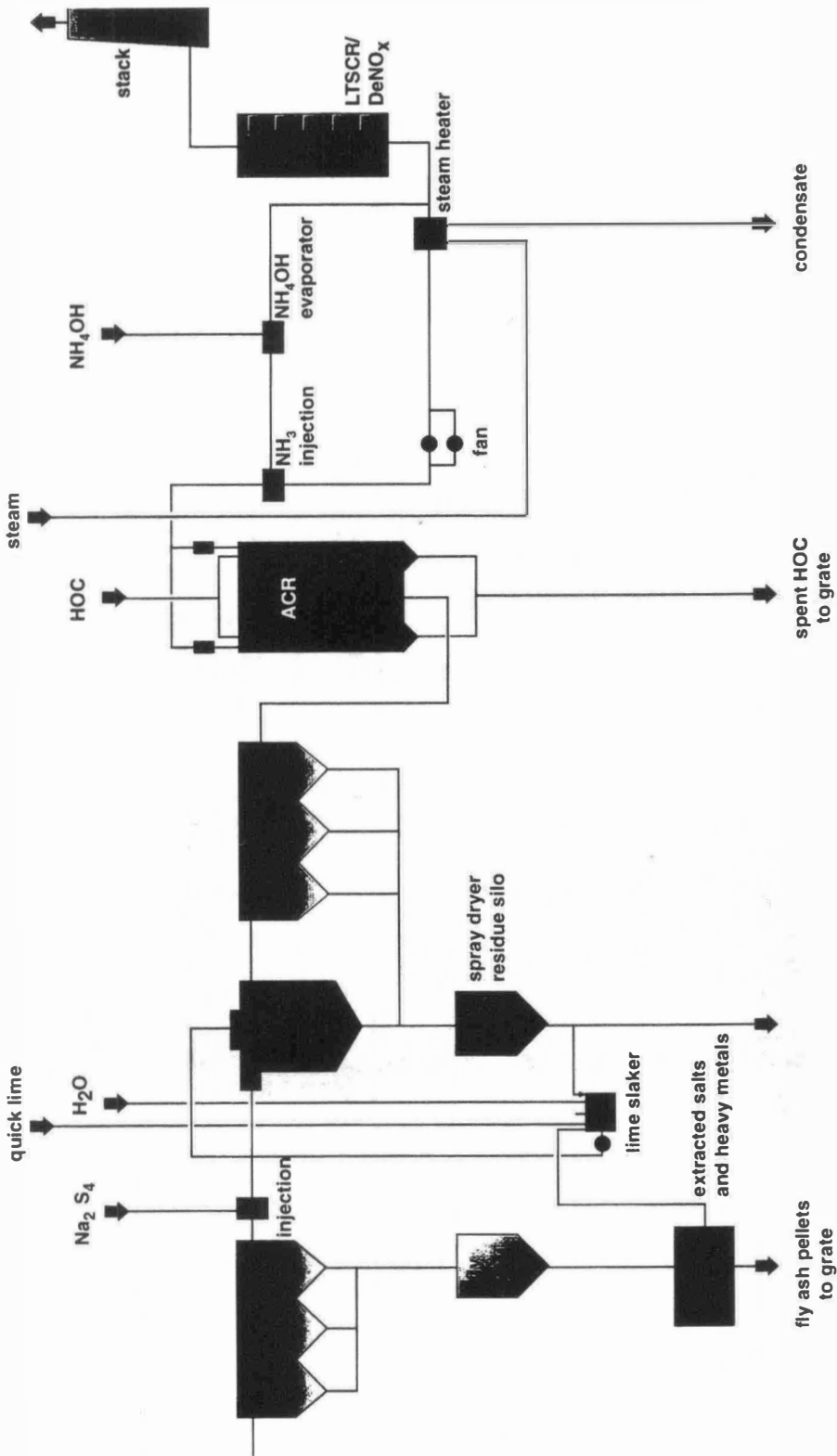
FIG. 7 17. BImSchV Retrofit of an Existing ESP/Spray Dryer/ Baghouse System with an ACR/LTSCR Combination

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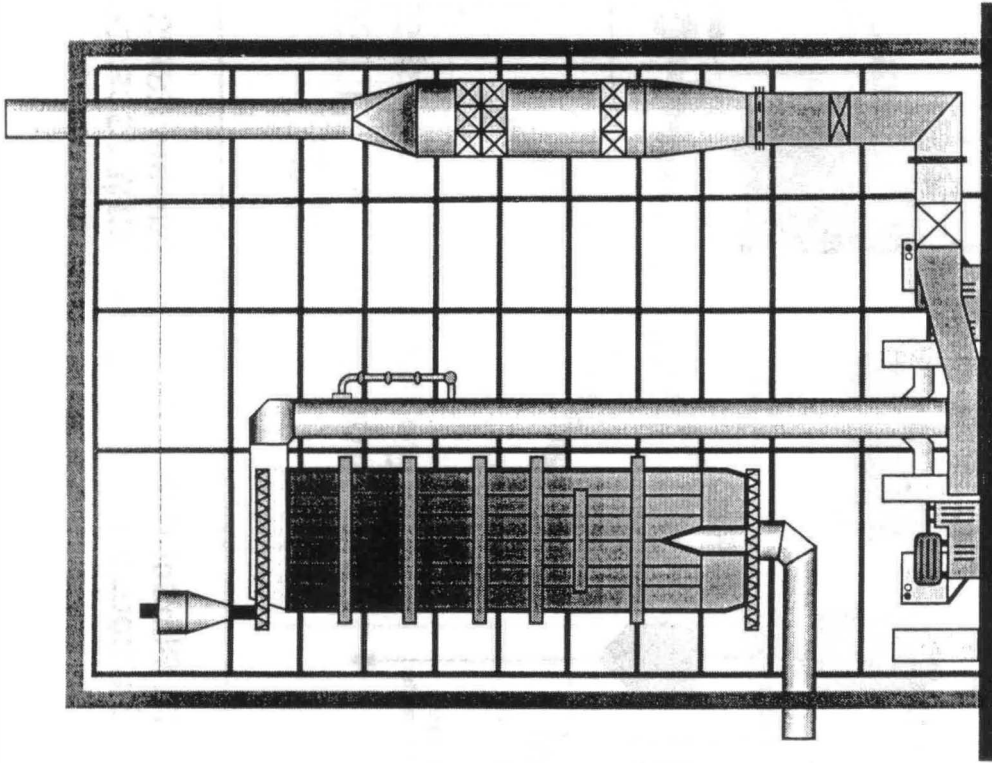
FIG. 8 General Layout of an ACR/LTSCR Combination for a 17. BImSchV-Retrofit of a MWC
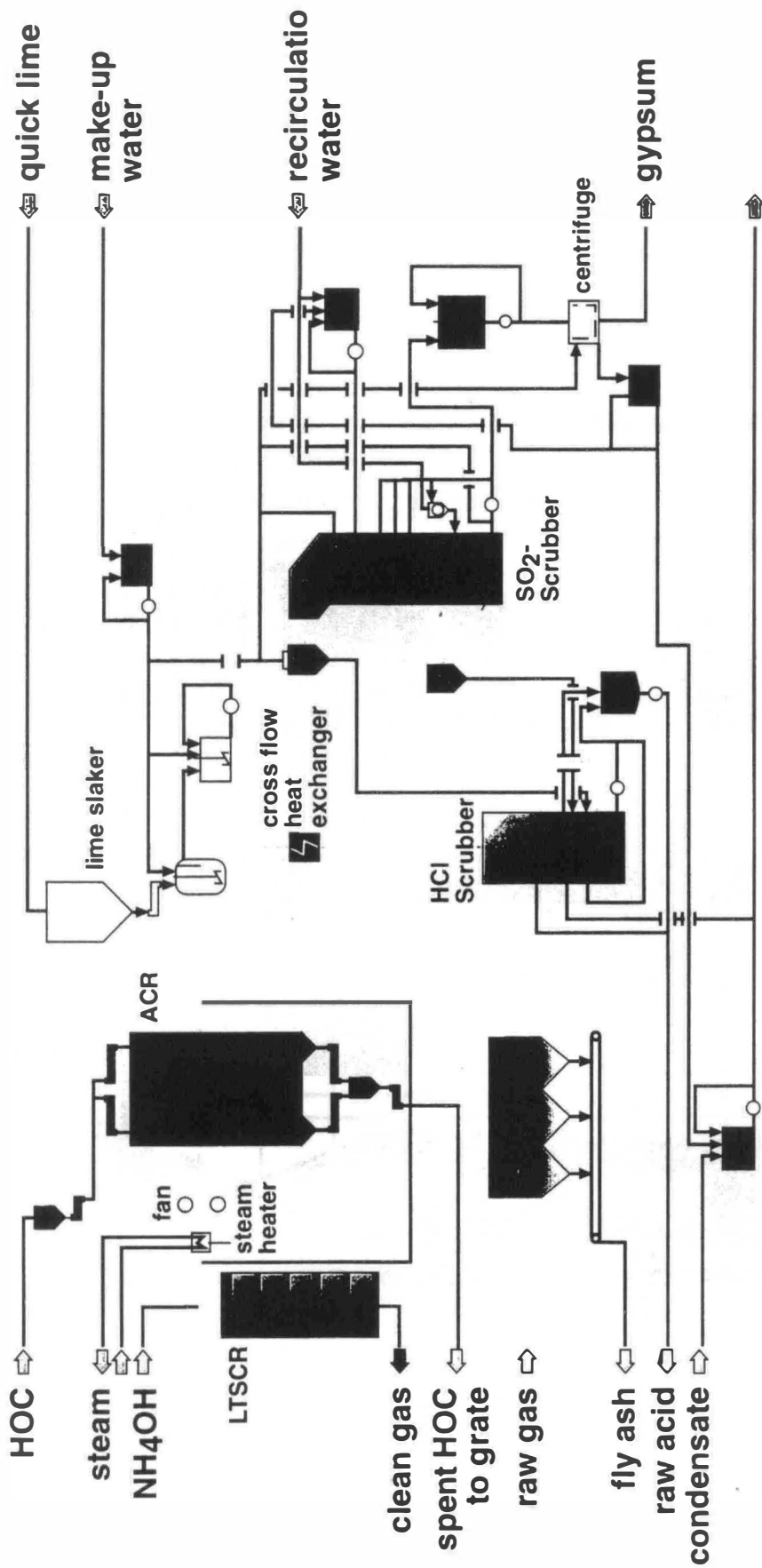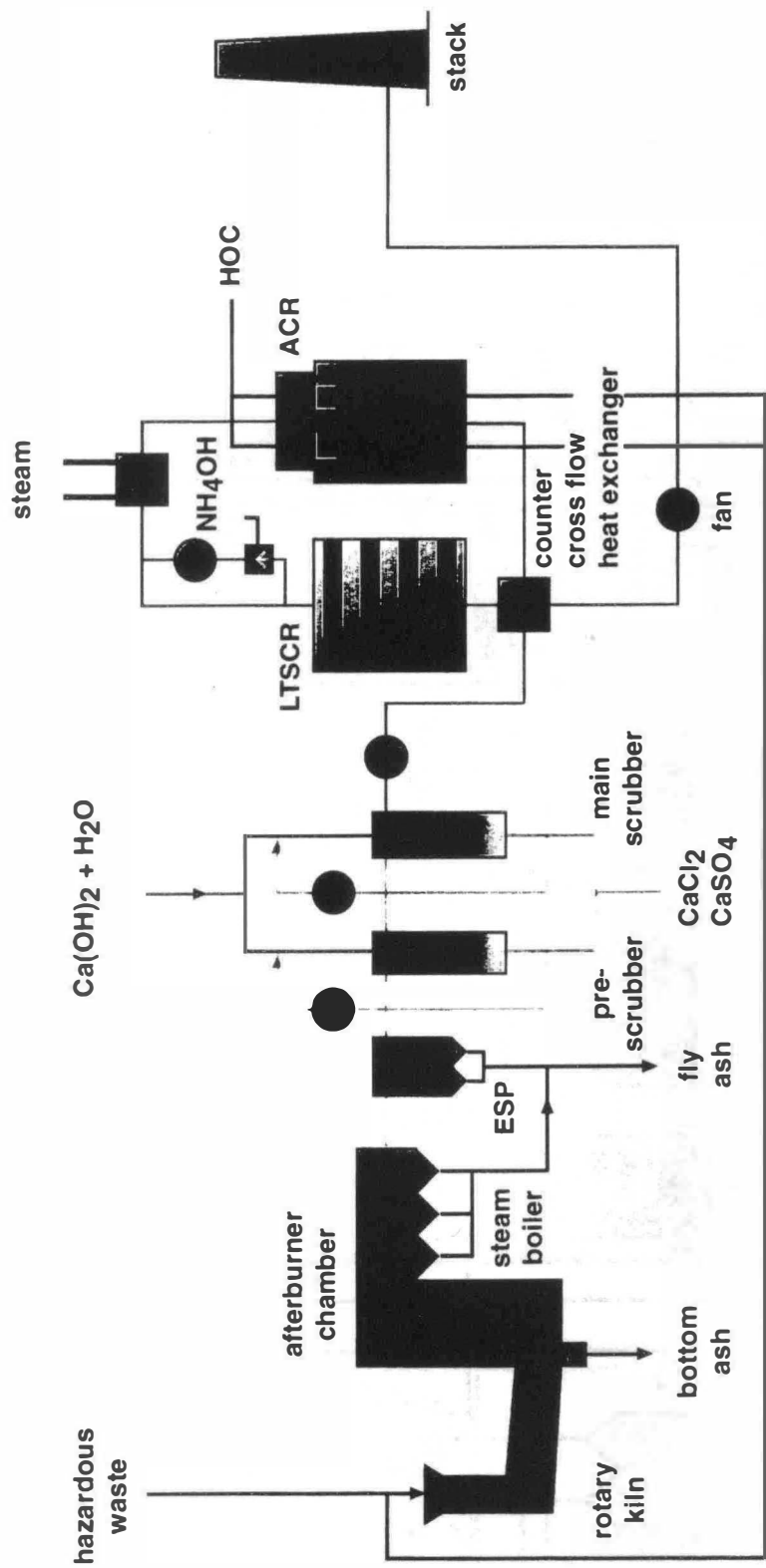
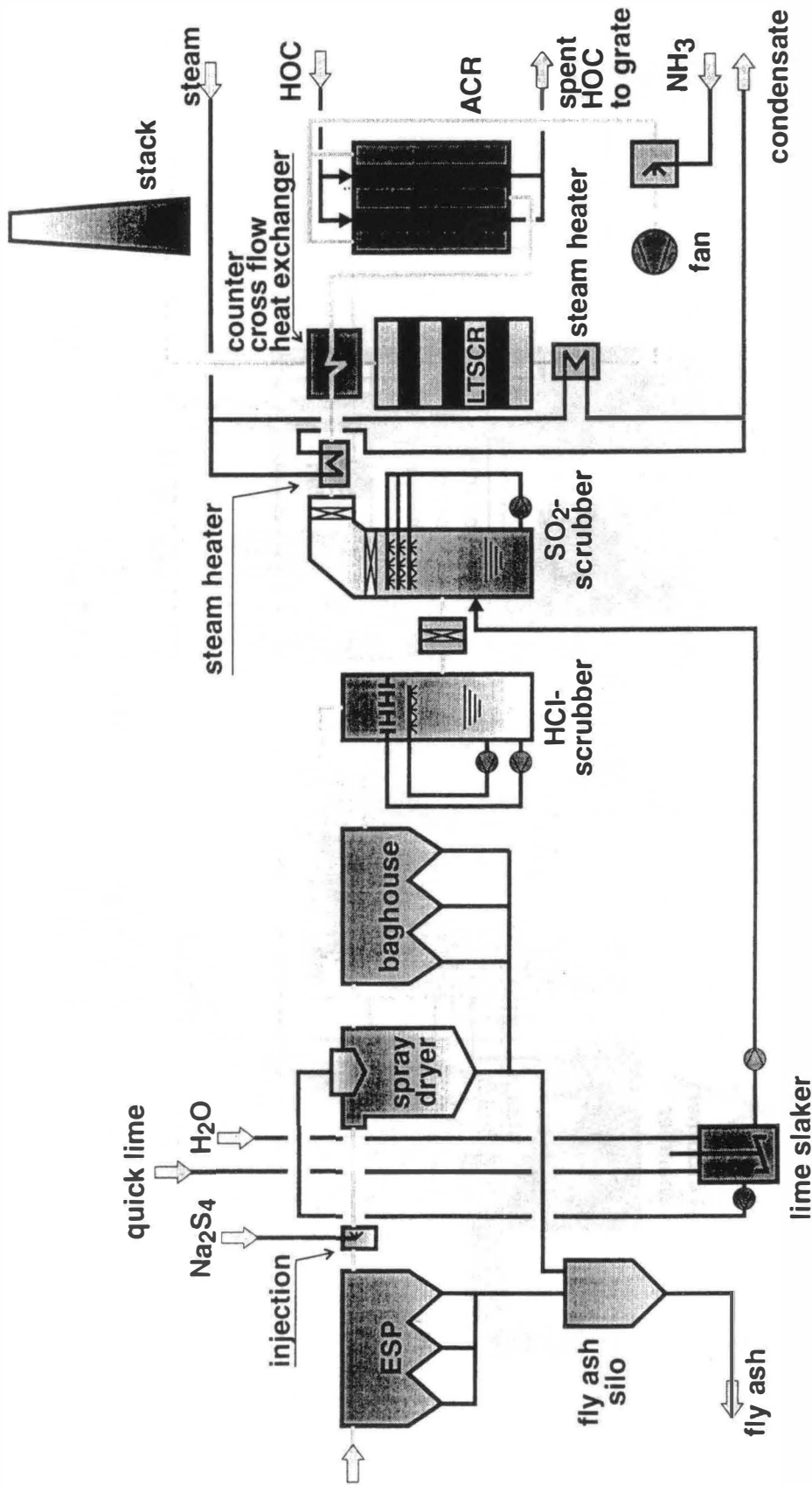**FIG. 9 Modern 5-Stage APC-Train of a MWC Including an ACR-LTSCR Combination**

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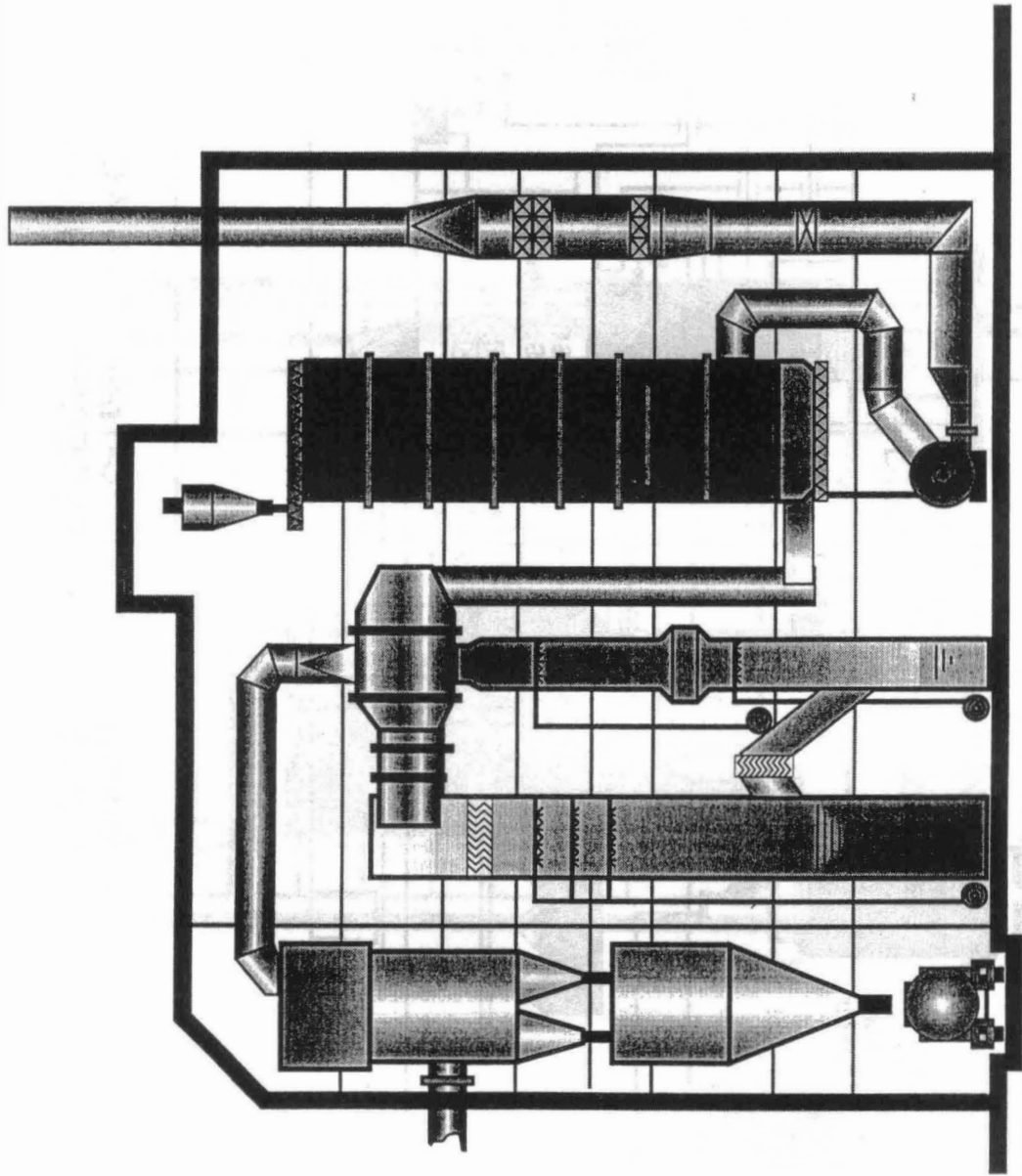
FIG. 10 General Arrangement of a Modern 5-Stage APC-Train
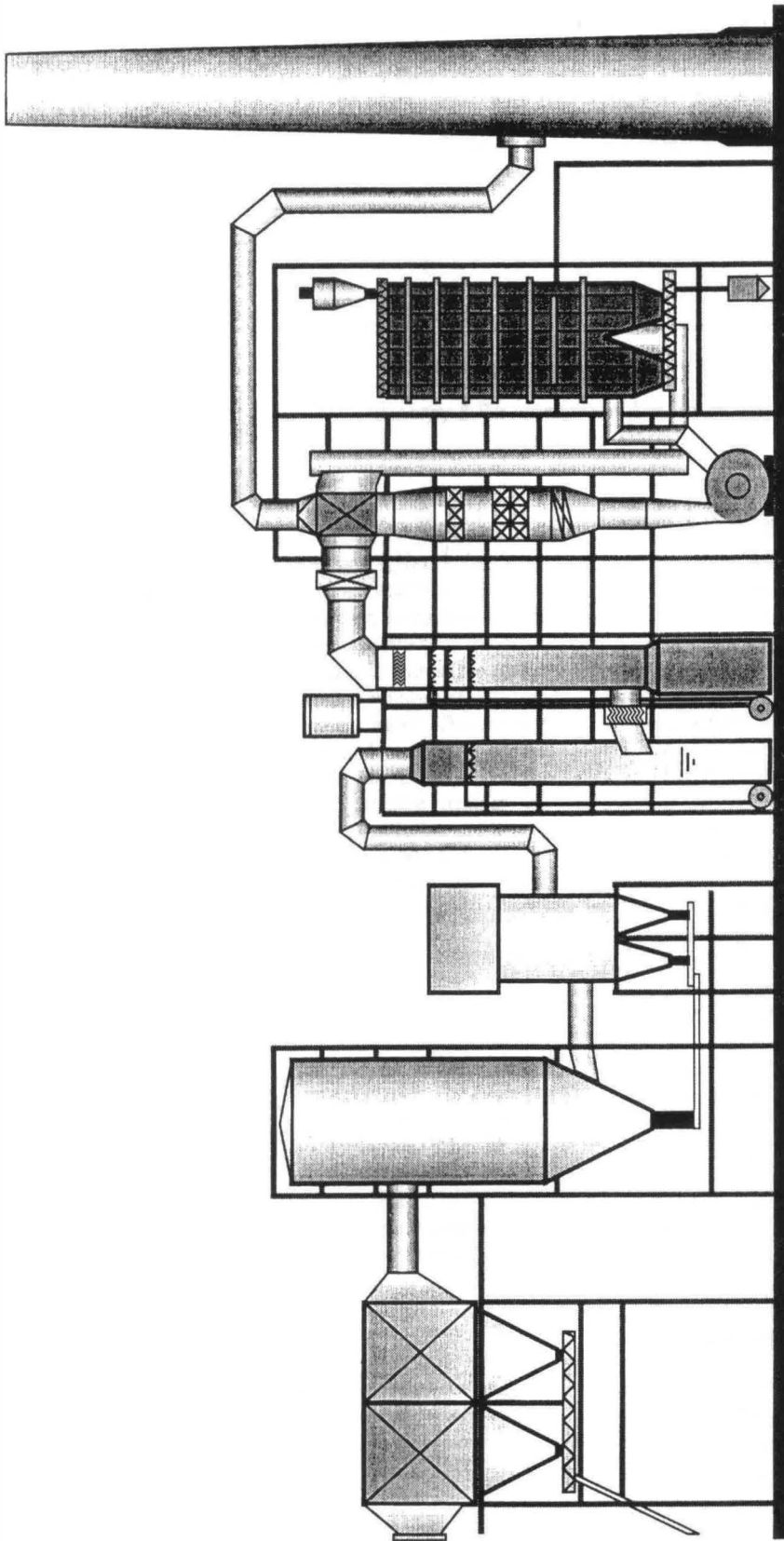With an Integrated ACR/LTSCR Combination for a MWC

FIG. 11 17. BImSchV-Retrofit of an Existing APC-Train of a HWC with an ACR/LTSCR Combination

FIG. 12   Modern Multi-Stage APC-Train of a HWC
Including an ACR/LTSCR Combination

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FIG. 13 General Arrangement of a Modern Multi-Stage APC-Train With an Integrated ACR/LTSCR Combination for a HWC