The Experience of SCR at Solnhofen and its Applicability to US Cement Plants

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Executive Summary

Solnhofer Portland Cement Works GmbH & Co. KG (Solnhofen) located in Solnhofen, Germany operates a preheater cement kiln, rated at 1800 MTPD (1980 STPD) clinker production.[1] In contrast, the cement kilns in Midlothian, Texas are either long wet kilns or modern preheater kilns with precalciners (PH/PC). The Midlothian PH/PC kilns are currently operating at approximately 5 times (TXI No.5) and 3.3 times (Holcim Nos. 1&2) the clinker production rate of the Solnhofen kiln. The raw materials for this facility consist of clay and waste stone. The waste stone is trucked to the facility from eleven different marble and granite operations owned by the same parent company as the cement plant.

The cement plant is equipped with both SNCR and SCR systems for NOx control. The SNCR and SCR systems do not operate simultaneously. According to the Plant Manager, Mr. Gerd Sauter, there would be no NOx control benefit gained by operating both systems simultaneously.

The facility operated a slipstream pilot test unit in 1998 and 1999. The facility began construction of the full-scale SCR system in 2000. The full-scale SCR system came online in 2001. A continuous program of testing different catalysts and cleaning methods has continued since the unit came online. However, when the site visit occurred on May 4, 2006, the SCR system was not in operation and had not been for the previous three months. Its ammonia injection system was disconnected. Instead, the facility was operating the SNCR system. Mr. Sauter would not commit to when or if the SCR system might be brought back online.

The facility achieves its current NOx emissions rate using a combination of a low-NOx burner, alternative/waste fuels, and either SNCR or SCR. According to Mr. Sauter, the NOx emissions are reduced by approximately 40 percent from the baseline of 1500 – 1800 mg/Nm3 to a level of 800-1200 mg/Nm3 by the use of a low-NOx burner and alternative/waste fuels. The use of either SNCR or SCR in addition to the low-NOx burner and waste fuels further reduces the NOx emissions by approximately 50 percent from the level achieved with the burner and waste fuels. With all three NOx controls operating (low-NOx burner, waste fuels, and either SCR or SNCR), the facility complies with the current NOx emission limit of 500 mg/Nm3. This represents approximately a 70 percent reduction from the uncontrolled baseline when the combination of technologies is implemented. <u>It is important to note that the emission limit of 500 mg NOx / Nm3 is achieved whenever either SNCR or SCR is used.</u> It is also important to note that there have been no long-term periods where NOx emissions have been maintained at levels significantly below 500 mg/Nm3. Therefore, both SNCR and SCR demonstrate NOx control efficiency of 50 percent at the Solnhofen facility. Due to variations in the NOx

[1] A short ton equals 2000 pounds. A metric ton equals 1000 kg or 2200 pounds.

concentrations at different kilns, the maximum possible control efficiencies will vary with the NOx concentration.

Introduction

This report has been prepared on behalf of the Portland Cement Association (PCA) to address questions that have been raised regarding the use of selective catalytic reduction (SCR) to control NOx emissions at the Solnhofer Portland Cement Works GmbH & Co. KG (Solnhofen) facility located in Solnhofen, Germany. For example, in a Draft Final Report prepared for the Texas Commission on Environmental Quality (TCEQ) by ERG, Inc., it was represented that the Solnhofen facility was achieving a NOx control efficiency of 80 percent through the use of SCR. The Draft Final Report also stated that Solnhofen's SCR system is achieving NOx emissions reductions "far in excess of those achievable using SNCR" and that the plant has been achieving approximately 200 mg NOx/Nm3.[2]

On May 4, 2006, Mr. Mark Terry and Mr. Frank Ruoss of Polysius; Mr. Randy Jones, Mr. Randy Walser, Mr. Brian Klotz, Mr. Bill Brown, and Mr. Greg Knapp of TXI Operations, LP; and Ms. Christa Russell and Mr. Robert Schreiber of Schreiber Yonley & Associates, visited the Solnhofen facility for the purpose of determining the design, efficiency and operational requirements of the facility's SCR system. This contingent of cement industry representatives met with the Solnhofen plant manager, Mr. Gerd Sauter, who provided the group extensive information regarding the facility equipment, raw materials, fuels, production rates, the SCR and SNCR systems, and past and current regulatory compliance and permitting. According to Mr. Sauter, no environmental group representative, academician, or US governmental representative has ever visited the Solnhofen facility.

This report first documents the plant information, then documents the regulatory history and the developmental history of the NOx emissions control systems as provided by Mr. Sauter. Then the report compares the Solnhofen facility to cement facilities in the United States.

Solnhofen Facility Description

The Solnhofen facility consists of one preheater cement kiln with a rated capacity of 1800 metric tons per day clinker. However, due to reduced demand for cement in Germany, the facility has reduced production to 1100 metric tons per day. The facility has had to modify the clinker cooler in order to efficiently operate at this reduced capacity. The facility raw materials are clay and waste stone. The waste stone is trucked to the plant from eleven different marble and granite facilities, which are owned by the same parent company as the cement plant. Some of this waste stone contains natural sources of ammonia. The raw materials are naturally low in both sulfur and alkali. Due to the low alkali, there is no alkali bypass. There is no alkali in the process to balance sulfur or chlorine in the raw materials or fuels. Therefore, the facility must limit the sulfur and chlorine in the fuels. The facility currently produces three types of cement.

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[2] ERG, Inc., "Assessment of NOx Emissions Reduction Strategies for Cement Kilns – Ellis County, Draft Final Report", Section 4.1.1 (December 19, 2005).

The facility currently burns Bunker C No. 6 oil and automobile shred fluff. The ratio is about 60:40. The fluff is a waste generated from the manufacturing of new automobiles. It is delivered to the facility in walking floor trailers. The facility utilizes a just-in-time delivery system, and has a maximum of 3 days storage onsite. The oil contains about 1% sulfur, and the chlorine content from all fuels is limited to about 0.2%. This limit is necessary to limit buildup of deposits in the preheater tower. The facility is currently permitted to burn bituminous coal, but has been unable to find an economic source of pulverized coal. The facility is not currently equipped with a coal mill. In the past the facility has utilized high carbon fly ash at the burner as well as animal meal as alternative fuels.

The facility controls particulate emissions in the kiln gas using a baghouse. Clinker cooler emissions are controlled with an electrostatic precipitator.

The facility air permit restricts the emissions of NOx from the kiln to 500 mg/Nm3, the emissions of particulate to 16 mg/Nm3, and the emissions of SO_2 to 50mg/Nm3. All limits are on a 24-hour average. Due to the lack of sulfur in the raw materials and the self-imposed low sulfur limit on the fuels, the emissions of SO_2 are nearly at the detection limit of the monitor. Opacity is not regulated in Germany either at the stack or as post-stack emissions. Condensable particulate emissions from the facility are not regulated and have not been measured. The facility has continuous stack monitors for NOx, SO_2, CO, O_2, NH_3, HCl, VOC, Hg, and stack gas volume, velocity, and temperature. The facility is required to conduct stack tests for dioxin/furan two to three times per year.

Regulatory History

According to Mr. Sauter, the facility entered into an agreement with the regulatory agency to install SCR and to meet a limit of 500mg/Nm3.

According to a review of the facility's permits, in 1990 the facility permit contained a NOx emission limit of 950 mg/Nm3. In 1995, an SNCR system was permitted with an emission limit of 850mg/Nm3. In 2000 the SCR construction permit states a goal of 200mg/Nm3. Mr. Sauter was insistent that the Solnhofen facility has not achieved a NOx emissions level of 200 mg/Nm3, and does not believe that he could achieve an emission rate that low. When the facility operating permit was issued in 2002, the NOx limit was set at 500 mg/Nm3, which remains the facility limit.

In accordance with the facility's agreement with the agency, the facility installed and operated a slipstream pilot test SCR system. The pilot equipment was installed and began testing in 1998 and continued testing in 1999. In 1999, the facility decided to go forward with the construction of a full-scale system. The construction of the full-scale system began in 2000 and was completed and the SCR system came online in 2001.

SCR at Other Cement Plants

Several other sources of information have stated that pilot testing was conducted in at least three other cement plants in Europe either prior to or concurrent with the pilot testing at Solnhofen.

Mr. Sauter confirmed that he is also aware of three tests. He confirmed information previously obtained from ELEX, a Swiss engineering company. ELEX first pilot tested SCR on a cement plant in Italy in 1996. Mr. Sauter indicated that the information for the testing in Italy is sketchy, but that it was his understanding that the test was not very successful. Then ELEX took their pilot SCR unit to Kirchdorf, in Austria. Although papers presented in 2001 describe this pilot test as successful, no documentation has been found to indicate that a full-scale unit was ever installed. After Kirchdorf, ELEX moved their pilot unit to Slite Cement in Sweden. Once again, the pilot test was successful, but the facility was able to achieve an extremely low emission rate using SNCR and a full scale SCR unit was not constructed. Mr. Sauter confirmed that the Solnhofen plant is the only existing full scale SCR on a cement plant. Mr. Sauter also confirmed that ELEX is currently constructing a full scale SCR at Cementeria Di Monselice in Bergamo, Italy. According to Mr. Sauter, the Monselice facility design has been copied from the current configuration at Solnhofen, and that the Monselice facility is very similar to Solnhofen in size and raw materials. ELEX has confirmed that the unit is expected to go online by early June 2006.

SCR at Solnhofen

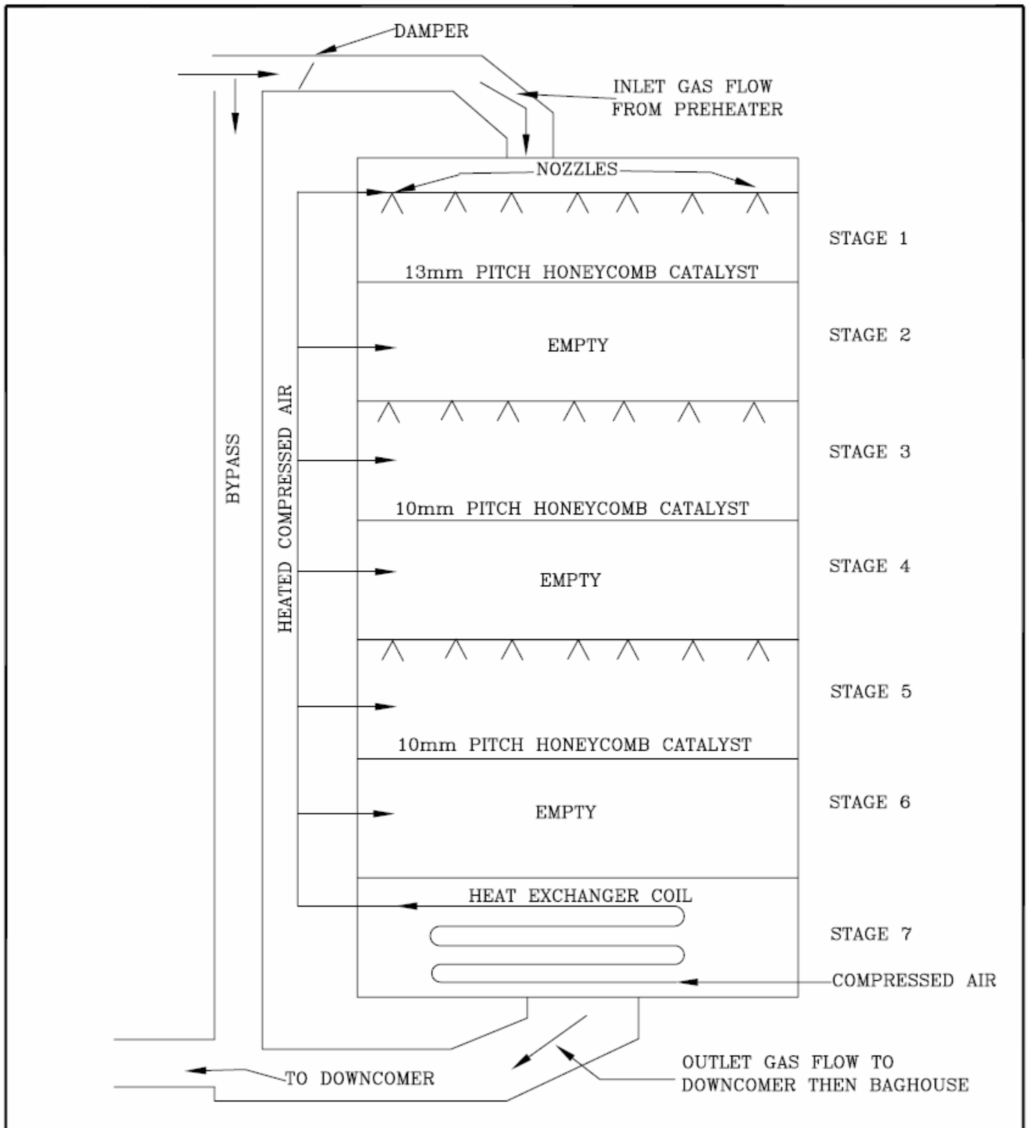
The slipstream pilot test unit at Solnhofen originally tested various plate catalysts. However, the dust loading in the preheater gas, 80-100 mg/Nm^3, and the pressurized cleaning eroded the catalyst from the metal substrate such that within 1000 hours the reactivity of the catalyst was no longer acceptable. The facility noted that, as the catalyst activity decreases, the ammonia slip increases.

When the full-scale SCR was constructed, honeycomb catalyst was installed, as it was believed that it would be more resistant to erosion. At that time, an 8 mm catalyst pitch was the largest opening commercially available. In this high-dust application, this catalyst plugged within minutes of beginning gas flow to the SCR. Several different options to clean the catalyst were tried. These included reversing the direction of the gas flow in the catalyst. Initially the gas would enter at the top of the SCR and flow downward, and then be switched to enter the bottom of the SCR and flow upward. It was theorized that this would result in cleaning the catalyst and that, when the pressure drop across the catalyst reached a set point, the flow could be reversed; therefore, continuous operation would be possible. This approach failed. Even if the lower catalyst did not plug quickly, the pore size and thickness of the catalyst beds did not allow for enough pressure to clean the catalyst by reversing the gas flow. Other cleaning methods were tried with limited success. Eventually the facility was able to have the catalyst vendor custom manufacture honeycomb catalysts with larger pitch and with various catalyst formulations. These other types were tested. The configuration of catalyst that has shown the best results to date is as follows:

1. The first SCR layer contains honeycomb catalyst with a 13 mm pitch.
2. The second layer is empty.
3. The third layer contains honeycomb catalyst with 10 mm pitch.
4. The fourth layer is empty.
5. The fifth layer contains honeycomb catalyst with 10 mm pitch.
6. The sixth layer is empty.

7. The seventh layer contains a heat exchanger, which preheats the air utilized for pressurized cleaning.

FIGURE 1 Solnhofen SCR Reactor sited next to preheater tower on right



SOLNHOFEN SCR SETUP				FIGURE 2 / APPROX. SCALE: NONE	SCHREIBER & YONLEY ASSOCIATES — ENVIRONMENTAL ENGINEERS
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Each catalyst bed contains six modules. Each of these modules contains 144 catalyst elements in a 12 X 12 arrangement. The total depth of each catalyst layer is 35.4 inches.

A bypass duct allows the facility to bypass the SCR whenever temperature or pressure drop requires a bypass.

When operating the SCR, the facility injects a 25 percent by weight ammonia in water solution, not at the inlet to the SCR as is typical of power plant applications, but rather in the preheater between the second and third cyclone in an area where the gases are at approximately 550 degrees C. The facility believes that this location allows for the best distribution of the NH_3 in the gas and therefore the most efficient SCR operation with the lowest ammonia slip. A separate location is utilized for the SNCR injection system. For the SNCR, the ammonia is injected into the preheater at a location where the gas temperature is approximately 1000 degrees C so that there is adequate temperature to drive the NOx reduction reaction without requiring a catalyst.

FIGURE 3 SNCR ammonia injections.

The most recent catalyst cleaning system utilizes preheated compressed air that continuously cleans the catalyst. Dry compressed air of about 900m3/ hr at 10 bar pressure passes through a heat exchanger coil that is located in the seventh stage of the SCR reactor, and then passes through insulated lines to each stage/layer of the reactor. Then the air passes into the reactor where a series of nozzles are located on two parallel bars, which span the width of the reactor.

The full cross-sectional area of the catalyst is reached by the cleaning system. Hydraulics is used to move the bar back and forth across the catalyst bed. The speed of the traverse is controlled electronically and can be adjusted when the pressure drop across the reactor begins to rise. The system cycles every 20 minutes, and cleans the catalyst in sequence. Roughly every 3000-4000 hours the facility bypasses the SCR to perform additional, more thorough cleaning. This unique cleaning system took 2 to 3 years to conceptualize, construct, and modify in order to achieve an effective method of catalyst cleaning for this particular cement plant's design and dust characteristics.

FIGURE 4 Compressed air system

Although the facility has managed to find an SCR setup that it considers to be reasonably reliable, they continue to explore options for improved operation and cost. The plant manager indicated that he has been attempting to find a catalyst manufacturer to produce a plate catalyst with a formulation that he believes will have more durability. Plate catalysts have less plugging issues and therefore less intense cleaning. However, to work effectively, the amount of reactive catalyst area must meet the NOx inlet loading as well as account for the expected amount of plugging. For plate catalyst this means finding the correct number of plates. For honeycomb, this means finding the optimum pitch size to minimize plugging while maintaining adequate reactive area. As the pitch size increases, the active area of catalyst decreases. Plate catalyst is less costly and easier to change out, and has less plugging issues. Honeycomb catalyst is more expensive and more difficult to change out.

The facility design includes large garage doors at each level, which can be fully opened to allow total access to the catalyst bed. The top of each layer is equipped with rails, which can be used to hoist catalyst modules from the reactor and lower them to the ground where the catalyst elements are removed and replaced, and then the module is hoisted back into position.

The plant manager has indicated that the most recent honeycomb catalyst has lasted 40,000 hours. The facility has spent around 3.1 million euros on the SCR system thus far. Mr. Sauter indicated that if he were to construct the facility again with what he has learned, the facility would probably cost about 2.5 million euros. These costs are based upon Solnhofen's site-specific conditions. The operating costs for the facility were not available.

NOx Control Efficiency

Mr. Sauter provided information about NOx emissions. He indicated that when burning only conventional fuel, fuel oil, or coal, the facility NOx emission rate ranges from 1500 – 1800 mg/Nm3, which is the uncontrolled baseline emission rate. With the introduction of the low-NOx burner and waste fuels, at the 60:40 blend of oil to fluff, the NOx emissions range from 800-1200 mg/Nm3; which is approximately a 40% reduction from the uncontrolled baseline. By using either SNCR or SCR in addition to the low-NOx burner and the waste fuels, the facility NOx emissions comply with the 500 mg/Nm3 limit contained in the permit. Therefore, the addition of either SNCR or SCR results in an additional reduction of approximately 50% below that achieved from the low NOx burner and waste fuels. Therefore, the NOx control efficiency demonstrated at Solnhofen is 50%. This is equivalent to the 50% reduction demonstrated using SNCR. Therefore, the overall reduction achieved by the combination of the low-NOx burner, waste fuels and either SNCR or SCR is roughly 70 % below the uncontrolled baseline levels.

It is important to remember that the chemical reaction, which takes place between ammonia and NOx, is the same for both SNCR and SCR. These reactions are:

$$4\ NO + 4\ NH_3 + O_2 \rightarrow 4\ N_2 + 6\ H_2O$$
$$4\ NH_3 + 2\ NO_2 + O_2 \longrightarrow 3\ N_2 + 6\ H_2O$$

The SNCR reaction occurs at approximately 900 – 1,000 degrees C (1,650 – 1,830 degrees F), while the SCR reaction occurs at a lower temperature, which is 300 – 450 degrees C (570- 840 degrees F). The chemical reactions are identical. The difference is that the SCR catalyst promotes the reaction by hosting a site upon the catalyst for the reaction (exchange of electrons between the individual atoms of the molecule) to occur. The catalyst therefore allows the reaction to take place at a lower temperature. It does not change the chemical reaction itself.

There has been no long-term operation of SCR that resulted in emission rates significantly lower than the permitted limit. The facility has established an operational NOx emissions set point for the facility, which is below the 500 mg/Nm3 limit. As a result, the overall average NOx emissions are between 475 and 500 mg/Nm3.

Mr. Sauter was asked whether he believed that the estimated achievable SCR control efficiency contained in the ERG, Inc. Draft Final Report for the Midlothian cement kilns (i.e., 80-85%) is actually achievable. He said no. His plant has only documented long term efficiencies in the range of 50%.

The facility monitors ammonia in the stack and has a very low ammonia slip in the range of less than 5 mg/Nm3 in compound operation (raw mill on).

Current Status

When the facility was visited on May 4, 2006, the SCR system was disconnected and not operating. The SNCR system was in use. Mr. Sauter stated that the SCR system had not operated for the past three months, and SNCR was being used instead. He would not commit to a date when the SCR system might be returned to use.

He is currently developing comparisons of the operational costs for the two systems. The SNCR system utilizes more ammonia; he is currently trying to determine how much more. The SCR system uses less ammonia, but requires the use of the compressed air cleaning system and the costs associated with the periodic replacement of catalyst (purchase of new and disposal of spent).

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FIGURE 5 Disconnected SCR ammonia injection system

Comparison with US Cement Plants

The Solnhofen cement plant is different from nearly all cement plants built in the United States after 1990, as follows:

1. It is an old preheater kiln. Most newer kilns are preheater kilns with precalciners (PH/PC). Many of the pre-1990 kilns in the US are long wet or dry kilns. Since 1999, nearly all PH/PC kilns permitted in the US are low NOx precalciner kilns.
2. The Solnhofen kiln is much smaller than the existing PH/PC kilns at Midlothian. Solnhofen is operating at 1100 MTPD (1210 STPD). The TXI Midlothian kiln is rated and operating at approximately five times this capacity and the Holcim-Midlothian kilns are rated and operating at approximately 3.3 times the Solnhofen capacity.
3. Due to market conditions in Germany, the Solnhofen facility has been operating at much lower than its rated capacity of 1800 tonnes/day. As a result of this and some concurrent changes in the cyclones, the facility did not have to increase fan size to install the SCR reactor. This would not be the case with most US modifications. The demand for cement in the US is very high and most plants are operating at or near capacity, and would not typically have the additional fan capacity necessary for the SCR reactor and the related equipment. Therefore, many of the US plants would have the additional cost of installation of a new fan if SCR were installed.
4. According to the Plant Manager, the Solnhofen facility raw materials contain minimal sulfur and alkali. The vast majority of US plants have sulfur and alkali. The fuel and raw material chemistry at Solnhofen is very different than that found in most US plants and would have minimal competing reactions. This allows the ammonia reaction with the NOx to be more efficient. It also means that the Solnhofen facility does not need an alkali bypass. Without sulfur in the raw materials, the Solnhofen facility does not need to be concerned with SO_2 to SO_3 conversion and therefore acid corrosions or detached plumes.
5. The lack of sulfur and alkali at the Solnhofen facility means that the dust particles reaching the SCR are not “sticky” and therefore are readily removed from the catalyst with compressed air. The oil used as fuel at Solnhofen has a maximum sulfur content of one percent. In contrast, most US cement plants, and particularly those in Midlothian, have significant sulfur and alkali in the raw materials as well as sulfur in the fuel. Due to the significant sulfur and alkali found in the raw materials and fuel used at the Midlothian kilns, Mr. Sauter indicated that catalyst beds installed in the Midlothian kilns would be harder to clean than the catalyst bed at Solnhofen.
6. The dust loading to the Solnhofen SCR ranges from 80-100 g/Nm3. The dust loading at TXI’s Midlothian kiln No.5 is 102 g/Nm3 and the dust loading at Holcim’s Midlothian kilns 1&2 are 177.8 and 139.5 g/Nm3 respectively. Mr. Sauter stated that, due to the differences in size, raw materials, and dust loading, a pilot test would be necessary before attempting to design a full-scale SCR for those facilities. See Figure 6

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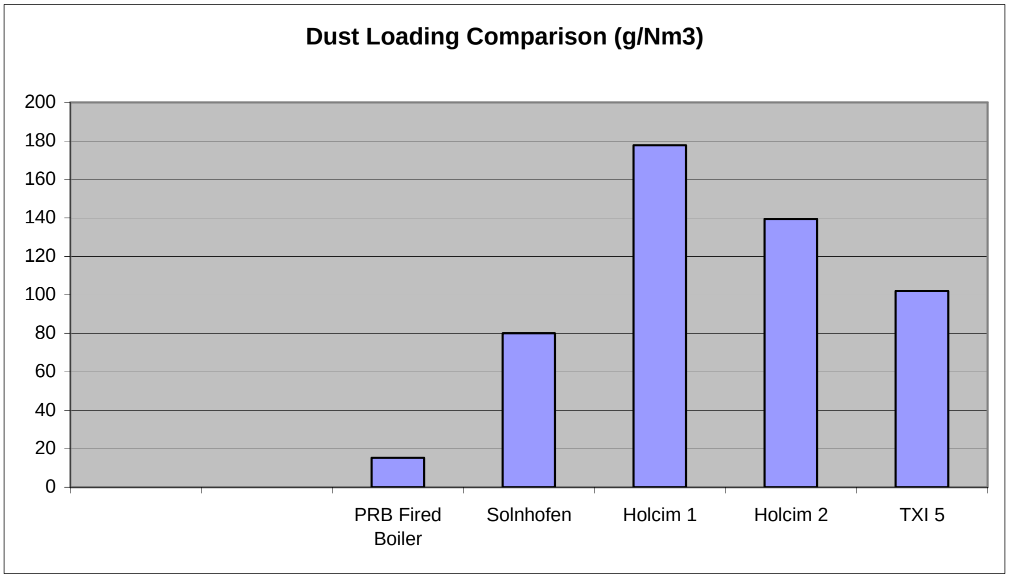
FIGURE 6 Dust Loading Comparison



7. Unlike US facilities, Solnhofen has no opacity limit, and would not have a regulatory compliance problem if a detached plume were to occur.
8. The concentration of NOx available for reaction is a factor in the efficiency of the reaction of the NOx with ammonia, with or without a catalyst. At high concentrations of a compound in a gas stream, the probablility that the reactant will contact the compound and react with it is high. At lower concentrations of a compound in a gas stream, the probability that the compound and reactant will come in contact is reduced. Therefore, at lower NOx concentrations, the efficiency of the reaction of ammonia will be lower, resulting in a lower control efficiency. A kiln with a NOx emission rate of 1500 mg/Nm3 can achieve a higher control efficiency than a kiln with a NOx emission rate at 500 mg/Nm3. This is important for kilns, such as those at Midlothian, where the NOx concentration in the preheater gases is already at a reduced concentration. The NOx emission rate for TXI Kiln No. 5, without the use of SNCR or SCR, is already below the emission rate at the Solnhofen kiln when it is using either SNCR or SCR. Therefore, it cannot be assumed that the addition of either of these technologies to a kiln such as the TXI kiln will result in the same control efficiency as that experienced at Solnhofen.

Conclusion

The Solnhofen cement plant began operating a full-scale SCR system in 2001. Since that time, the facility has been trying different catalyst design, size, and formulas, and has had to address plugging problems. Various mechanisms for cleaning the catalysts have been tested, and the facility ultimately designed their own site-specific system for continuous cleaning of the catalyst beds when the SCR is in operation. The plant manager still hopes to find a plate catalyst that is erosion-resistant enough that he can replace the honeycomb catalyst with plate. Therefore, although the facility has been using the full-scale SCR since 2001, it has essentially been performing ongoing testing, and is not yet at the hoped for final design configuration.

The facility also has an SNCR system in place. Prior to the SCR system shutdown, whenever the SCR system has been bypassed for extended periods of time, the facility utilizes the SNCR system. They do not use both the SNCR and SCR concurrently, and Mr. Sauter indicated that no benefit would be expected from doing so. When used at the Solnhofen facility, the SNCR reduces the NOx emissions to the same level as that achieved by the SCR system. The facility also utilizes waste fuels for about 40 percent of the fuel input. The use of these waste fuels and a low-NOx burner results in a reduction of the NOx emissions of approximately 40 percent below baseline uncontrolled emissions, prior to and without the use of either the SCR or SNCR systems. When SNCR or SCR are used in combination with the low-NOx burner and the waste fuels are used, an additional NOx reduction of about 50 percent has been achieved. This constitutes a total NOx reduction below uncontrolled baseline emissions of approximately 70 percent. Both SNCR and SCR achieve a control efficiency of about 50 percent below the inlet NOx concentration. Therefore, the SNCR system is as effective at controlling NOx at Solnhofen as the SCR system. The plant manager at Solnhofen stated that, based upon his experience, the 80-85 percent NOx control efficiency estimated by ERG, Inc. in the Draft Final Report for SCR at the Midlothian cement kilns is not achievable. He also was emphatic that slipstream pilot testing is necessary prior to design and operation of SCR at other cement plants. The Solnhofen design is not readily transferable to other kilns.

Solnhofen is not currently using the SCR system, and is achieving the same level of control with the SNCR system. The plant manager is currently comparing the operational costs of SNCR and SCR. He did not commit to a time when the SCR system might be brought back on-line. And if it is, he hopes to make further changes to the catalyst.

Based upon all of the information provided by the plant manager of Solnhofen, and knowledge of US cement plants, it is clear that the SCR technology developed at Solnhofen is still in a state of development. Based on the significant differences between that plant and typical PH/PC plants in the US, the technology is not readily transferable to US operations. At a minimum, slipstream pilot testing must be conducted before attempting to design and construct a full-scale system. Both the plant manager at Solnhofen and the catalyst manufacturers contacted in the US have been emphatic on the need for the pilot testing. At Solnhofen the SNCR system achieves the same level of NOx reduction as that achieved by SCR. The capital cost for the SCR system is significantly higher than the cost of the SNCR system, and the same level of control achieved. No information is currently available about the comparison of the operating costs for the two systems.