

PHOTO-CATALYTIC CONSTRUCTION MATERIALS
AND REDUCTION IN AIR POLLUTANTS

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March 2006

This report was produced with the help of a 2004-2005 contract from the California State University Faculty Research Fellows Program for the California Senate. The opinions expressed in this report do not necessarily reflect the opinion of the California Assembly Speaker's Office. The Coordinator of the Faculty Research Fellows program is Professor Jim Cox, Center for California Studies, California State University, Sacramento. For information on the Faculty Research Fellows program and a list of all previous reports, visit http://www.csus.edu/calst/Government_Affairs/faculty_fellows_program.html

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EXECUTIVE SUMMARY

The objective of this study was to review and evaluate the available results of existing research on the use of photocatalysts for reduction of air pollutants, focusing primarily on the data from the European Union project titled “Photo-catalytic Innovative Coverings Applications for De-pollution Assessment (PICADA)”.

This study indicated that construction material containing or coated by TiO₂ mineral can oxidize pollutants adsorbed on their surfaces when exposed to sunlight. The pollutants evaluated ranged from certain dye material to volatile organic compounds (VOCs) and nitrogen oxides (NO_x). Under the controlled experimental conditions in a laboratory environment, a dye adsorbed on the TiO₂-containing material was oxidized effectively, indicating the feasibility of having “self-cleaning” material on building facades that can eliminate discolorations and stains. Extremely limited laboratory studies with few VOCs showed the potentially successful control of VOCs in air by using TiO₂-coated surfaces. A single pilot test also showed that through the use of sufficient TiO₂ material on building facades significant reduction in NO_x can be achieved.

Yet, there seems to be numerous questions left unanswered by the current research. A systematic approach at the laboratory scale is required to answer many questions regarding the reaction rates, mechanisms, reaction products (especially potentially hazardous chemicals, such as ozone and organic radicals), the fate of reaction products, types of pollutants, (various VOCs,) and the effect of various variables, including TiO₂ particle size, the type of TiO₂, the percent of TiO₂ in the mixture, the thickness of the mixture required (penetration distance of reactions), temperature, humidity, and concentration of pollutants. For both indoor and outdoor applications, the reaction products need to be identified and quantified under various conditions to make sure no hazardous chemicals will be released as a result of these photocatalytic reactions. In addition, the useful life of the TiO₂ - containing material needs to be determined. The effectiveness of TiO₂ in colored matrix should be evaluated, assuming that painting all the buildings in white would be impractical. These laboratory experiments should be conducted in a fully-controlled system that can operate under steady-state and continuous-flow conditions (as opposed to the batch lab experiments used in earlier research) to be able to simulate realistic conditions. A conceptual model of the process describing the mass transport and reaction of various pollutants under various realistic scenarios for outdoor atmospheric conditions should be developed. The sensitivity of the model to all possible variables and atmospheric conditions should be evaluated.

The results of the laboratory studies together with the modeling efforts should provide a better understanding of the process for its potential use in air pollution control. It is recommended that this systematic approach should be followed before any consideration of a full or even a pilot scale implementation of the process.

INTRODUCTION

Photocatalysis is a reaction mechanism involving the interaction of a semiconductor catalyst, e.g., TiO_2 with light energy. Semiconductors are characterized by a narrow band gap energy between their valence and the conduction bands. The band gap of TiO_2 is 3.2 eV^1 . The absorption of a quantity of light energy greater than or the same as the band gap of the semiconductor results in an abrupt transfer of electrons from the valence to the conduction band and the consequent creation of a hole in the valence band and an electron that can move freely in the conduction band. When the hole reacts with water, a hydroxyl radical (OH^\bullet), which is a strong oxidant, is produced. A conduction band electron on the other hand is a strong reducer; hence, the species adsorbed onto the surface of the TiO_2 can go through oxidation or reduction by OH^\bullet or the electron, depending upon the chemical characteristics of the adsorbed species. TiO_2 can form two different crystallographic forms: rutile and anatase. The photocatalytic activity of anatase is generally much higher than the rutile form.

Photocatalysis has received a wide academic interest over the last three decades because of its potential to be used for pollution control in air and water²⁻⁹. This also promoted industrial applications and private research. A huge increase in worldwide patent applications was observed and in 1999 more than 400 international applications were filed¹⁰.

Photocatalysis has been used to disinfect water, and oxidize numerous organic chemicals in water into non-hazardous by-products. Among the air pollutants, nitrogen oxides (NO_x), hydrocarbons and organic chlorides have received the most attention, primarily because the reactions in the atmosphere between NO_x and several hydrocarbons

in the atmosphere under solar irradiation is known to create the condition referred to as Photochemical Smog—a mixture of chemicals and oxidants of health hazards within heavy traffic urban areas. In addition, sulfur oxides (SO_x), formaldehyde, ammonia, chloroform, gasoline components, e.g., benzene, toluene, and odors from tobacco and have been subjects of research by photocatalytic radiation for their removal.

Many in Japan studied the feasibility of using TiO_2 as part of white wall painting for indoor pollution control in conjunction with visible lighting or UV lamps to cure the “sick building syndrome” resulting from insulated and airtight buildings. These so called air purifiers were devised for home- and office in small sizes of about $100\text{m}^3/\text{h}$ of air flow rate to traffic tunnel ventilation systems in sizes as large as $1,500,000\text{m}^3/\text{h}$. These systems are usually combined with a filter or electrostatic precipitator to remove both hazardous gases and air-borne particulate matter. Another application tried in Japan is to coat the construction materials with the photocatalyst and remove air pollutants around the buildings and structures¹¹. This so-called “passive air purification” is claimed to clean the ambient air under the sunshine, while saving energy and labor^{11, 12}.

TiO_2 was used as part of white cement¹³ and other construction material. The main reason was to maintain the aesthetic characteristics, in particular the color of the surface over time due to continuous removal of stains and dirt by the attack of OH^\bullet generated through the photocatalytic activity of TiO_2 . Other “smart” construction materials and coatings that capture and decontaminate air pollutants are being developed in Europe. In the presence of sunlight, plaster, mortar, concrete, and coatings containing TiO_2 are expected to eliminate the pollutants. The process involves the pollutant gases and organic compounds to diffuse through the porous surface and stick to the TiO_2 nano-

particles of the construction materials and coatings. Absorption of UV light by the incorporated TiO₂ leads to its photo-activation and formation of OH^{*}, which degrades the pollutants absorbed onto the particles. It is reported that the oxidation products created by this process, e.g., organic acids, are washed away by rain and/or neutralized by alkaline calcium carbonate contained in the materials.

It is reported that after 7,000 square meters of road surface in Milan, Italy were covered with a photo-catalytic cement material, there was up to 60 percent reduction in the concentration of nitrogen oxides¹⁴. However, there is no additional information to help with evaluation of this claim.

The performance of such materials is currently being tested by a European consortium of private companies, research institutions, and the European Commission's Joint Research Centre (JRC) as part of a program for innovative construction materials to help in the fight against air pollution.

OBJECTIVES OF THE PROJECT

The major objective of this project was to review of the available results of existing research on the use of photocatalysts for reduction of air pollutants. The data from the Photo-catalytic Innovative Coverings Applications for De-pollution Assessment (PICADA) project of the European Union was to be gathered and analyzed in addition to the other related work. The data assessment was to include the effectiveness of the construction material using TiO₂, the extend of the pollution reduction, and the transport mechanisms and the fate of TiO₂ after being introduced into the environment. In addition, the cost of addition of TiO₂ to construction material was to be estimated. Finally, based on best available information, a recommendation was to be provided for the State of

California whether to consider incorporating TiO₂ into exterior and interior construction material.

PROCEDURE

A thorough literature search was conducted to include two books and numerous refereed publications, conference papers, reports, magazine articles, and web-sites (See the Reference List). In addition, several attempts have been made to contact several PICADA officers, including Mr. Andre Piavaux, the general coordinator of the project. Because of unfortunate passing away of Mr. Piavaux, attempts to gather data from PICADA research were delayed considerably until late August while attending an International Conference in Stasbourg, France, I was able to contact Mr. C. Gobin of GTM Construction, a partner in the PICADA Project. Subsequently, in mid-September 2005 four publications on PICADA project results were sent to me by Mr. Gobin's co-worker, Ms. H. Andre.

REVIEW OF THE PICADA RESEARCH RESULTS

The web-site of the Photo-catalytic Innovative Coverings Applications for Depollution Assessment (PICADA) Project and numerous other non-refereed papers and web-based literature indicate that researchers are actively studying photo-catalytic activities of new materials and coatings. The location of the Indoortron test facility is in Ispra, Italy, and this is a facility of the Physical and Chemical Exposure Unit of the JRC's Institute for Health and Consumer Protection. The new materials and coatings are being developed as part of the PICADA project, with the expectations to tackle the persistent

problem of urban smog and to help meet the European Union target of reducing NOx levels to under 21 parts per billion per year by 2010¹⁵.

The TiO₂-based smart construction materials are being tested in laboratory settings under varying experimental conditions of humidity, temperature and UV radiation, to simulate a real world setting. The project has started on January 1, 2002 and was expected to finish in 2005. However, at this point it is obvious that the project has not met the target date. The entire project was expected to cost U.S. \$4.2 million, and the European Commission committed to funding the project up to U.S. \$2.34 million.

According to the telephone conversations on 8/23/2005 with Mr. C. Gabin of GTM Construction, a French partner in the PICADA Project and the person who is currently the manager of the project, the feasibility of the process in terms of reducing NOx concentrations in air and keeping the surface of the material that is coated with TiO₂ has been verified in laboratory settings. However, taking the process to the level of field scale is hampered primarily because of varying weather conditions, e.g., wind speed, wind direction, intensity of sunlight and angle of sunrays, as well as for lacking a conceptual mathematical model of the process. As opposed to the controlled experimental conditions in laboratory settings, comparison of field-scale results under the varying conditions would be quite difficult. Furthermore, Mr. Gabin stated that even the mathematical modeling of the process would be quite complicated because of numerous factors and parameters that are difficult to incorporate into the model.

It should be noted that none of the four papers on the PICADA Project forwarded by Mr. Gabin's Office is a refereed publication. Furthermore, two of the publications describe lab-scale experiments, only one describes a pilot-scale experiment.

The first paper is a Press Release¹⁶, which provides an overview of the objective of using “smart” construction materials containing TiO₂ as part of the PICADA project, i.e., to reduce levels of NO_x gases and other toxic substances, such as benzene in air, and to keep the materials clean. There is no scientific or technical information provided in this paper. The paper includes a list of the participating partners, from France, Italy, Denmark and Greece.

The second paper, which was presented at the CIB World Building Conference in 2004, primarily concentrates on self-cleaning ability of the novel construction materials that contain TiO₂¹⁷. In this laboratory study, TiO₂ nanoparticles (20 nm average size) in an anhydrous phase were mixed with cement, lime, sand and water, and was directly cast in 10 cm Petri dishes to expose only the upper surface to the surrounding air. A second material created by mixing TiO₂ particles with cement and some fillers was applied as a 1 mm thick paint on mortar in 10 cm Petri dishes. Both materials contained the same amount of TiO₂ and were cured under the same temperature and humidity and storage time. An organic dye (Rhodamine B, a polycyclic aromatic hydrocarbon, PAH) was sprayed on both surfaces and then exposed to UV light in the same wavelength of natural sunlight. At an intensity of 3700 Lux, the dye degraded by 65% within 24 hours on both surfaces. This result is an indication of the oxidative capabilities of the photocatalytic process, and its potential success to keep building facades coated with TiO₂ clean. It does not allow anyone to extrapolate the conditions to real life pollution cases, under different weather and lighting conditions. Furthermore, these results do not indicate anything about air pollution control capability of the process.

In a specially designed testing chamber in the laboratory (1.5 L size), a mixture of benzene, toluene, ethylbenzene, and o-xylene (BTEX), which represented pollution by VOC (volatile organic compounds) was irradiated in a catalytic chamber that contained the two previously described TiO₂-materials at a distance of 25 cm from the radiation source. The UV intensity in the chamber was measured as 12.4 W/m². Significantly higher decomposition of the VOCs was estimated in terms of VOC destruction rate (μg/m²-hr) divided by VOC concentration in μg/m³). The results came out as about 1-2 m/hr for TiO₂-materials versus negligible rate for non TiO₂-containing mortar. If we try to apply this rate to formaldehyde, a cancer-causing air pollutant, and if we assume that air contains 1,000 μg/m³ (about 1.5 ppm at standard temperature and pressure), it can be estimated that a surface area of about 0.25 m² of air covered with TiO₂ will be needed for each m³ of air to reduce this concentration to 500 μg/m³ in an hour. This required surface area will be only 0.04 m² if 6 hours of sunlight is assumed to strike the surface. Again, these results are indications of potentially successful control of VOCs in air by using TiO₂-coated surfaces. However, the results are specific to the conditions tested, and cannot be extrapolated to real life cases without additional information, as will be discussed later in this report.

The researchers used the same test chamber to measure the NO₂ removal rate in air. It was observed that NO₂ adsorbed on TiO₂ test material during dark phase, and upon radiation, it was oxidized to NO₃⁻, which was recovered by rinsing the TiO₂ material. Both construction test materials showed 3-4% NO₂ destruction efficiency over the control with no TiO₂. These results indicate that NO₂ adsorbed on building material during the night can undergo photocatalytic oxidation when exposed to light the following day.

However, these extremely limited results do not allow extrapolations to estimate how much NO₂ can be removed from air and how much TiO₂ material is needed to have any significant reduction in NO₂ under realistic conditions.

The third and fourth papers were authored by the same individuals and presented at the Urban Air Quality Conference, Valencia, Spain in 2005. The third paper¹⁸ described a laboratory study in two different systems used to oxidize NO and NO₂ (NO_x). The first system was a 450 L size test glass chamber illuminated by a 300 W lamp mounted outside the chamber. Two types of TiO₂-containing materials were tested: a mineral paint and an organic-based coating. The mineral paint, which was primarily ordinary Portland Cement with some organic additives, and obtained from Italcementi Co., was supplemented with 3% by weight TiO₂. The second material was a translucent water-based organic with siloxanes used as binders. It was obtained from Millenium Chemicals, Inc. This material was supplemented with 10 or 5% TiO₂ sol-gel in Wacker BS45. These materials were embedded in Petri dishes so that only the upper surfaces were exposed. The dishes were placed 50 cm below the lamp. The temperature and humidity were controlled by placing the chamber in a stainless steel room. The second system was 30 m³ stainless steel environmental chamber with temperature and humidity control and fans to mix the air. A 300 W lamp was placed at 1.5 m from the center of four TiO₂ samples, each embedded on a 1 m² glass surface. The UV intensity varied from 2.1 to 4.6 W/m², depending upon the location. For both systems, NO values were measured continuously in air by an NO_x analyzer. Both systems were operated as batch, and this makes the extrapolation of the results to real life conditions difficult without integration of the results into a comprehensive mathematical model of the process.

Both systems showed significant removal of NO and NO₂ by catalytic effect of TiO₂ under the experimental conditions. It was observed that NO degraded 20 to 100% faster than NO₂. The same photo-degradation rates were obtained with the mineral paint with 3% TiO₂ in both test systems. However, the translucent organic-based coating produced a little higher rate in the lab system with 10% TiO₂, and much lower rate in the macro system with 5% TiO₂ than the mineral paint. Furthermore, the organic based coating material produced ozone as a reaction product. Oxidation of NO also produced NO₂, which is another undesirable end product.

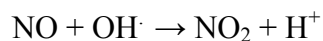
The fourth paper¹⁹, which was also presented at the Urban Air Quality Conference in Spain in 2005, describes a pilot test conducted at the CTG cement plant in Guerville, France. The purpose was to estimate the photocatalytic efficiency of a TiO₂-containing mortar. The test site consists of three mini-canyon streets with a width of 2 m, length of 18.2 m, and the wall height of 5.2 m., produced at a scale of 1:5. The walls of the first canyon were bare. The walls of the second canyon were covered with TiO₂-treated mineral paint from Italcementi, Co.(amount of TiO₂ not reported), and the walls of the third canyon were covered with the same material, except it did not contain TiO₂ (reference canyon). The street ground was treated with tar emulsions and gravel without any TiO₂. The test was conducted during in summer of 2004. From July 9-August 23, 2004, the pollution source, which was a perforated gas pipe distributor connected to an engine, was placed into the TiO₂ canyon. The pollution source was moved to the Reference Canyon during the testing between August 23 through September 3, 2004. The pollution source was operational between 9:00 am to 4:00 pm. The measured NO_x levels in the canyons varied with the time of the day and wind direction, as well as the

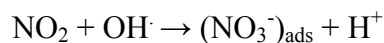
orientation of the canyon wall, as expected. However, overall the NO_x concentrations measured in the TiO₂ canyon was 40-80% lower than those measured in the Reference Canyon, on an average. The report indicates that precautions were taken in data analysis to avoid the possible errors caused by conducting the two sets of experiments at different time periods under different meteorological conditions and background values of NO_x. Hence, it can be concluded that through the use of sufficient TiO₂ material on building facades significant reduction in NO_x can be achieved. However, this study did not report the results on ozone formation.

COMMENTS ON EXISTING RESEARCH RESULTS

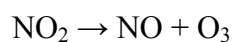
The following conclusions can be derived from this literature review:

1. Existing literature is very limited, especially for pilot or full-scale demonstration of the effectiveness of TiO₂-containing construction material in reducing air pollutants. It is extremely hard to extrapolate this limited information to real life conditions to estimate the extent of the reduction in air pollution that can be brought about by the use of TiO₂ as part of construction material.
2. Overall, there seem to be more anecdotal reports with limited available technical information than reliable and refereed publications on the process.
3. The process seems to be quite effective in keeping the material clean, i.e., oxidizing pollutants causing discolorations and stains on the material.
4. The laboratory tests on photo-catalytic degradation of NO_x provide conflicting information on the reaction by-products and reaction mechanisms. For example, Cassar et al., (13) report that the reaction mechanisms involve the following steps:





In other words, final reaction product is reported to be nitrate as adsorbed in the matrix of the construction material. Vallee, F. et al.¹⁷ agree with this conclusion. However, Anpo¹¹ reported formation of N₂, N₂O and O₂ during photocatalytic degradation of NO. Furthermore, Maggos et al.¹⁸ observed formation of ozone (O₃) during photocatalytic degradation of both NO and NO₂, and formation of NO during photocatalytic degradation of NO₂, which indicate reduction of NO₂ back to NO, as follows:



The reduction of NO₂ back to NO is quite plausible, since the electron hole on radiated TiO₂ surface can act as a reducing agent.

For pollutants other than NO_x, such as VOCs, there is basically no information in the accessible literature on the reaction by-products and reaction mechanisms.

5. Elucidation of the reaction mechanisms has paramount importance in avoiding the formation of hazardous chemicals, such as ozone and perhaps some organic free radicals resulting from VOCs, during the process. For example, in one experiment Maggos et al., reported the formation of up to 0.035 ppm of ozone during degradation of about 225 ppm of NO₂. This is quite troubling considering the fact that the 8-hr exposure standard of US EPA for ozone is 0.085 ppm in air.
6. With the exception of the pilot test reported by Maggos et al.¹⁹, lab scale experiments in batch systems are described. In these experiments, the concentration of the pollutants and the products vary with time, and eventually all

- NO and NO₂ disappear. Although these experiments show the effectiveness of the process in degrading these pollutants, they also produce limited information in terms of long-term effectiveness of TiO₂-containing materials, varying concentrations of pollutants and their interactions in a simulated environment. For example, since the pollutants adsorb on TiO₂-containing materials, when light is absent in night time and the pollutant concentrations also drop due to reduced transportation activity, the adsorbed pollutants may potentially desorb back.
7. It is not clear from the existing literature how long the TiO₂ pigments can continue to be functional. In other words, can reaction products and the chemicals that do adsorb, yet do not wash away from the surface reduce the effectiveness of TiO₂ over time?
 8. Nevertheless, it was shown by a pilot test that under the experimental conditions tested, construction material mixed with TiO₂ pigments can reduce NO_x by 40-80%. However, the experimental results were not verified under different atmospheric and other experimental conditions.
 9. The information if TiO₂ can effectively function in a colored (non-white) matrix is not readily available in the literature.
 10. Available information on the PICADA project and others do not provide a systematic approach to reveal the effects of a) TiO₂ particle size, b) the type of TiO₂ (anatase versus rutile), c) the percent of TiO₂ in the mixture, d) temperature, e) humidity, and f) concentration of pollutants on effectiveness of the TiO₂-containing materials.

11. Information on the effectiveness of TiO₂-containing materials on various components of smog (other than NO_x) is not available.

COMMENTS ON AVAILABILITY AND THE COST OF TIO₂:

The bulk price of TiO₂ powder on January 13, 2006 was about \$1.12 – 1.24 per pound in the US²⁰. The demand for TiO₂ has been increasing worldwide, according to the UK's Millenium Chemicals, the second largest TiO₂ producer in the world and one of the PICADA project partners. The new demand is arising from the plastics and coatings industries, especially in Asia/Pacific. Accordingly, the price is expected to go up and the delivery lead times will continue to grow longer.

The potential use of TiO₂ as part of construction materials will further increase the demand. Titanium is a mineral found in many locations, but primarily in several African countries and in Australia. The mining of titanium can produce acid and poison nearby water bodies, unless the acid mine drainage is controlled at the mining locations. The cost of environmental protection through application of proven approaches and technologies must be added to the overall cost of TiO₂.

ESTIMATION OF THE COST OF USING TIO₂ FOR REDUCING AIR POLLUTION

Because of the limited information on the process, it is very difficult to produce any meaningful cost estimation for air pollution reduction efforts using TiO₂.

However, if an attempt has to be made, the following major assumptions have to precede this attempt:

1. The results of the pilot scale experiments can be extrapolated to expect significant (40-80%) reduction in NO_x in air.

2. Basis of calculations: A street that is 10 m wide and 91 m long. The street is flanked on both sides by buildings with an average height of 26 m.
3. The surface area to be covered by TiO₂-containing material: 91 m x 26 m = 2,366 m².
4. The thickness of the TiO₂-containing material on the facades of the buildings: 1 mm.
5. TiO₂ is mixed with a cement mortar and paint mixture at 3% (by weight).
6. The cement mortar that TiO₂ is mixed with has a density of about 2.1 kg/m³.
7. The price of TiO₂ is \$1.12/lb
8. There is no difference between the labor costs of mixing cement with TiO₂ and applying the TiO₂ mixture on the surface of the buildings versus applying a regular surface material with no TiO₂.

Under these conditions, it is estimated that incorporating TiO₂ will only add about \$360 to the cost compared to the cost of applying a regular surface material.

If this cost can be further extrapolated to a urban area of about 10 square miles (with a street coverage equivalent to about 6,000 streets used as a basis in calculations above), the required amount of TiO₂ will be about 1,920,000 pounds at a price of about \$2.2 million. Obviously, the assumption here is that all the building facades will be covered with TiO₂-containing material!

No attempt has been made in this study to estimate the monetary benefit of pollution reduction due to TiO₂ use. This requires a fairly complex and multidisciplinary approach that should involve all the stake holders in this issue, and it will ultimately be a public policy decision.

RECOMMENDATIONS

The PICADA Project seems to be focusing on reducing outdoor NO_x by using TiO₂-containing construction material on exterior surfaces of buildings and relying on the UV waves of solar rays, whereas research in Japan seems to be focused primarily on decontaminating indoor air by using TiO₂-containing paint and artificial light.

The limited information available in the literature indicates that both approaches could be successful in reducing air pollution. Yet, there seems to be numerous questions left unanswered by the current research.

A systematic approach at the laboratory scale is required to answer many questions regarding the reaction rates, mechanisms, reaction products (especially ozone and organic radicals), the fate of reaction products, types of pollutants, (various VOCs,) and the effect of various variables, including TiO₂ particle size, the type of TiO₂, the percent of TiO₂ in the mixture, the thickness of the mixture required (penetration distance of reactions), temperature, humidity, and concentration of pollutants. For both indoor and outdoor applications, the reaction products need to be identified and quantified under various conditions to make sure no hazardous chemicals will be released as a result of these photocatalytic reactions. In addition, the useful life of the TiO₂ - containing material needs to be determined. The effectiveness of TiO₂ in colored matrix should be evaluated, assuming that painting all the buildings in white would be impractical. These laboratory experiments should be conducted in a fully-controlled system that can operate under steady-state and continuous-flow conditions (as opposed to the batch lab experiments used in earlier research) to be able to simulate realistic conditions.

A conceptual model of the process describing the mass transport and reaction of various pollutants under various realistic scenarios for outdoor atmospheric conditions should be developed. The sensitivity of the model to all possible variables and atmospheric conditions should be evaluated.

The results of the laboratory studies together with the modeling efforts should provide a better understanding of the process for its potential use in air pollution control. It is recommended that this systematic approach should be followed before any consideration of a full or even a pilot scale implementation of the process.

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