

## Full Length Article

# First evaluation of a multicomponent flue gas cleaning concept using chlorine dioxide gas – Experiments on chemistry and process performance

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## ABSTRACT

This work has investigated a multi-pollutant flue gas cleaning concept based on oxidation using chlorine dioxide (ClO<sub>2</sub>) gas with subsequent absorption. The chlorine dioxide gas converts the relatively insoluble nitric oxide (NO) to the more soluble nitrogen dioxide (NO<sub>2</sub>). This makes a downstream wet scrubbing process feasible for simultaneous removal of sulphur oxides (SO<sub>x</sub>) and nitrogen dioxide (NO<sub>2</sub>).

An experimental evaluation of the proposed process using chlorine dioxide gas has been performed on a laboratory scale. The experimental setup, designed and built by Akzo Nobel, consists of a reactor for oxidation, a flue gas condenser and a wet scrubber. The results show that ClO<sub>2</sub> gas oxidises NO with high efficiencies under a wide range of process conditions, also in the presence of sulphur dioxide (SO<sub>2</sub>). The more ClO<sub>2</sub> gas is added, the higher the degree of NO oxidation and the total nitrogen oxides (NO<sub>x</sub>) removal efficiency becomes. The results also show that the presence of water strongly increases the removal of SO<sub>2</sub>, which is believed to be an effect of liquid phase nitrogen-sulphur interactions. The absorption solution, sodium carbonate and sodium sulphite, is efficient in removing NO<sub>x</sub> (especially NO<sub>2</sub>) from the oxidised flue gas. The total NO<sub>x</sub> reduction at 0.6 ClO<sub>2</sub>:NO mole ratio and subsequent wet scrubbing is between 79% and 94%, depending on the process conditions used. The total SO<sub>2</sub> reduction in the scrubber is between 97% and 100% independent of ClO<sub>2</sub> gas addition. Furthermore, the total NO<sub>x</sub> balance shows that the major part of the NO<sub>x</sub> is converted to nitrate in the condensate liquor and as nitrite in the absorption solution. A higher ClO<sub>2</sub> gas addition and a higher reactor temperature convert more of the NO<sub>x</sub> to nitrite in the absorption solution.

## 1. Introduction

The combustion of fossil fuels in particular causes emissions of nitrogen oxides (NO<sub>x</sub>) and sulphur oxides (SO<sub>x</sub>) to the atmosphere. These gases are associated with adverse effects on human health and are responsible for environmental problems like photochemical smog, acid rain, tropospheric ozone and ozone layer depletion. With the concern over the environmental and health consequences of these pollutants, stricter legislation and regulations are continually being implemented, which requires improved emission control systems.

Today, emission control systems in a conventional combustion plant typically employ technologies designed to remove one specific pollutant; several emission control systems are then combined in series in order to meet the emission regulations. Selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR) are two conventional post-combustion emission control technologies for NO<sub>x</sub>

abatement, while flue gas desulphurisation (FGD) is the most common technology for SO<sub>x</sub> removal. Multi-pollutant technologies for the removal of two or more pollutants in a single stage have been and are being investigated; see for example Refs. [1–3]. Multi-pollutant technologies have, however, not yet achieved commercial breakthrough. The driving force behind the development of emission control systems is to improve the removal and cost efficiency, as well as to make implementation easier in retrofit cases in particular. The benefits mentioned for multi-pollutant systems are lower capital and operating costs than for a series of traditional systems to remove the same number of pollutants, and their footprint is often smaller in size than conventional single pollutant counterparts treating a similar volume of flue gas [4].

For the efficient removal of nitric oxide (NO) in a wet scrubber, there is a need to convert the relatively insoluble NO to soluble oxides like nitrogen dioxide (NO<sub>2</sub>), dinitrogen trioxide (N<sub>2</sub>O<sub>3</sub>) and dinitrogen tetroxide (N<sub>2</sub>O<sub>4</sub>). Oxidation chemicals like ozone (O<sub>3</sub>), hydrogen

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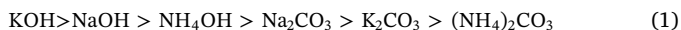
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**Nomenclature**

CO <sub>2</sub>	carbon dioxide
ClO <sub>2</sub>	chlorine dioxide
FGD	flue gas desulphurisation
HADS	hydroxylamine disulfonic acid
HCl	hydrochloric acid
H <sub>2</sub> O <sub>2</sub>	hydrogen peroxide
HNO <sub>3</sub>	nitric acid
HNO <sub>2</sub>	nitrous acid
H <sub>2</sub> SO <sub>4</sub>	sulphuric acid
K <sub>2</sub> CO <sub>3</sub>	potassium carbonate
KOH	potassium hydroxide
Na <sub>2</sub> CO <sub>3</sub>	sodium carbonate
NaOH	sodium hydroxide

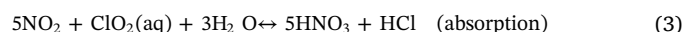
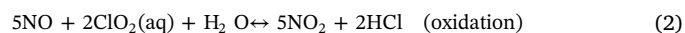
NaHCO <sub>3</sub>	sodium hydrogen carbonate
Na <sub>2</sub> SO <sub>3</sub>	sodium sulphite
NH <sub>4</sub> OH	ammonium hydroxide
(NH <sub>4</sub> ) <sub>2</sub> CO <sub>3</sub>	ammonium carbonate
NO <sub>2</sub>	nitric dioxide
NO <sub>x</sub>	nitric oxides
NO	nitric oxide
N <sub>2</sub> O	nitrous oxide
N <sub>2</sub> O <sub>3</sub>	dinitrogen trioxide
N <sub>2</sub> O <sub>4</sub>	dinitrogen tetroxide
O <sub>3</sub>	ozone
SCR	selective catalytic reduction
SNCR	selective non-catalytic reduction
SO <sub>2</sub>	sulphur dioxide
SO <sub>x</sub>	sulphur oxides

peroxide (H<sub>2</sub>O<sub>2</sub>) and chlorine dioxide (ClO<sub>2</sub>) can be used for this purpose [5–8]. The soluble oxides can then be absorbed in a subsequent absorber tower, forming nitrous acid (HNO<sub>2</sub>) and/or nitric acid (HNO<sub>3</sub>), depending on the degree of oxidation, i.e. the NO:NO<sub>2</sub> ratio, and on the type and composition of the absorber solution [9]. According to Kuropka [9] and references cited therein, the sequence of activities of alkaline solutions used for the absorption of nitrogen oxides diminish in the following order:



Jin et al. [7] and Deshwal et al. [8] used a laboratory bubbling reactor to evaluate a method for simultaneous removal of sulphur dioxide (SO<sub>2</sub>) and NO. In the proposed method an aqueous chlorine dioxide solution (ClO<sub>2</sub> gas dissolved in water) was used to conduct both oxidation and absorption processes in the bubbling reactor. ClO<sub>2</sub> oxidises NO to NO<sub>2</sub> according to Reaction (2), which can then form HNO<sub>3</sub> according to Reaction (3). Deshwal et al. [8] also mention that if SO<sub>2</sub> is present in the flue gas it can react with ClO<sub>2</sub> according to Reaction (4). The results obtained in these studies showed an SO<sub>2</sub> removal efficiency

of 100% and an NO<sub>x</sub> removal efficiency of 66–72% in optimised process conditions in the bubbling reactor [7,8].



Other studies using sodium hypochlorite (NaClO), sodium chlorite (NaClO<sub>2</sub>) and chloric acid (HClO<sub>3</sub>) in wet scrubbing processes have also shown promising NO<sub>x</sub> and SO<sub>x</sub> removal efficiencies [9–12].

Furthermore, in 2002, Patrikainen [13] developed a two-stage flue gas scrubber to decrease NO<sub>x</sub> emissions from pulp mill recovery boilers and showed that nearly 80% of the incoming NO in the flue gas can be removed by using this multi-stage flue gas scrubbing method. The technology involves an oxidation stage applying aqueous ClO<sub>2</sub> or NaClO for oxidation of NO to NO<sub>2</sub> and then a reducing stage using sodium sulphite (Na<sub>2</sub>SO<sub>3</sub>). It was concluded that at optimal process conditions (pH > 11 and Na<sub>2</sub>SO<sub>3</sub> concentration of about 100 g/L) in the reducing stage, 60% of the NO<sub>2</sub> was converted to N<sub>2</sub> and 20% to

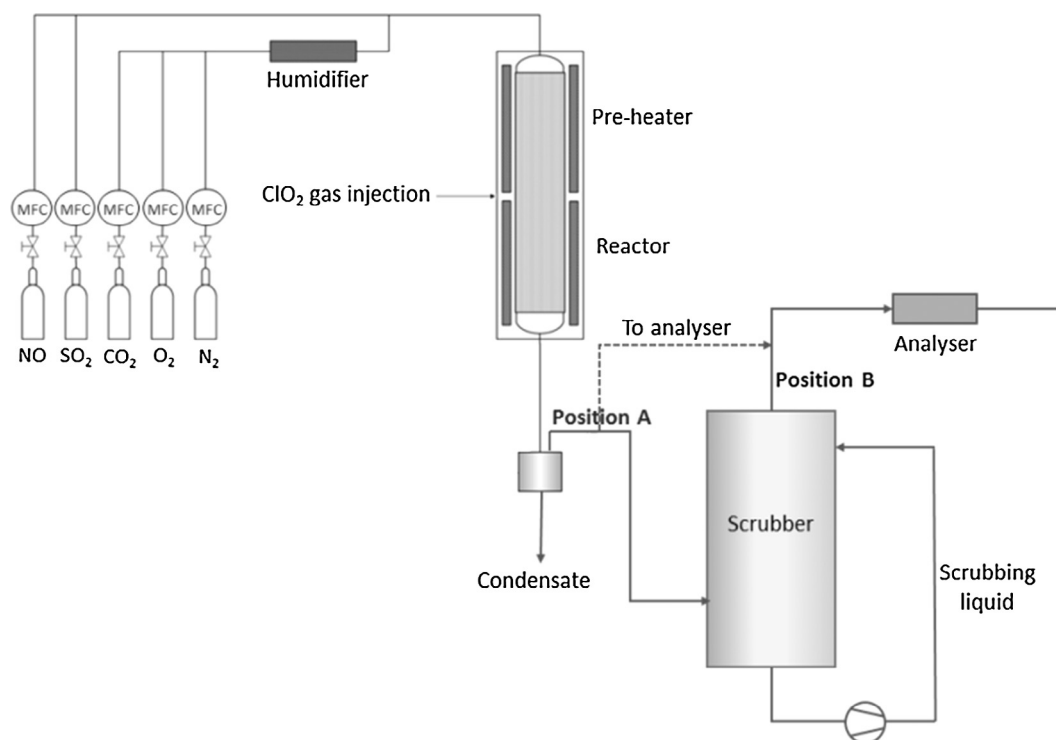


Fig. 1. A schematic description of the process outline.

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