Kinetic model for the design of chlorite-based wet oxidative scrubber for NO_x capture

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Nitrogen oxides (NO_x) are major air pollutants from combustion processes, contributing to air pollution and causing serious environmental and health issues. While NO_x emissions have decreased in Europe and North America due to stricter regulations, the challenges persist in various industrial sectors. Control strategies include combustion optimisation and direct flue-gas treatment, with common methods being Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR). Emerging technologies like Non-Thermal Plasma (NTP) and Wet Oxidative Scrubbers (WOS) are also gaining prominence, with WOS being an energy-efficient and ready-to-use technology, particularly suitable for retrofitting existing scrubbers. Oxidants like H_2O_2, $Na_2S_2O_8$, $NaClO_2$, and K_2FeO_4 are used to treat flue gas. Sodium chlorite ($NaClO_2$) is notable due to its stability, ease of handling, and high effectiveness under acidic conditions. Research indicates that $NaClO_2$ effectively oxidises NO_x, with its performance further enhanced by a low pH and the presence of SO_x in flue gas. These conditions promote Acidic Oxidation Mechanisms (AOM), accelerating mass transfer and significantly reducing the oxidant's dosage.

However, the lack of general design criteria for chlorite-WOS systems and the need to optimise oxidant dosage prompted us to investigate the fundamental aspects of the process, such as gas-liquid reaction kinetics, and more generally, to estimate the Enhancement Factors (E_L) that determine the efficiency of reactive absorption.

In this work, we experimentally estimated the E_L dataset for NO_x absorption in $NaClO_2$ solutions using a lab-scale falling-film absorber. The experimental activity was conducted by varying the main process parameters: temperature (20–50 °C); gas flow rate (0.1–0.51 m^3/h), liquid flow rate (21–50 L/h), sodium chlorite concentration (0.01–0.125% w/w), NO_x inlet concentration (100-1000 ppm_v), and absorbing solution pH (3-10). Experimental data were modelled with Danckwerts' kinetic equation for a pseudo m^{th}-n^{th}-order non-reversible reaction to estimate the kinetic parameters: pre-exponential reaction constant, activation energy, and reaction orders. The experimental results indicate that E_L increases with the operating dosage (d_{op} = molar flow rate of $NaClO_{2(aq)}$/molar flow rate of $NO_{x(g)}$, [mol/mol]), at fixed temperature and pH. Furthermore, increasing the gas temperature has only a slight effect on outlet NO_x concentrations; it significantly influences E_L by accelerating the reaction rate in the liquid phase. Additionally, lowering the solution pH from 10 to 3 results in up to a 50% increase in E_L, as the AOM becomes dominant. Under acidic conditions, chlorite dismutation leads to the formation of more reactive chlorine-based oxidants, which greatly accelerate the reaction kinetics.

Keywords: *Nitrogen oxide, Gas-Liquid reaction, Wet Oxidative Scrubber, Flue Gas Treatment*