Development of Nitric Oxide Oxidation Catalysts for the Fast SCR Reaction

In an effort to reduce catalyst costs in SCR installations, this project has focused on the development of catalysts which are active and selective for the oxidation of NO to NO₂. Two main reactions contribute to NOₓ conversion over ammonia SCR catalysts:

\[ 4\text{NH}_3 + 4\text{NO} + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O} \quad (1) \]
\[ 2\text{NH}_3 + \text{NO} + \text{NO}_2 \rightarrow 2\text{N}_2 + 3\text{H}_2\text{O} \quad (2). \]

NOₓ formed in combustion processes is typically composed of >90% NO and reaction (1) therefore dominates. In the case that equimolar amounts of NO and NO₂ are present, NOₓ reduction occurs according to equation (2). This is the so-called “fast” SCR reaction. Expressed in terms of a first order rate law, the ratio of the two rate constants, \( k(2)/k(1) \), is at least ten at \( T > 200 \, ^\circ\text{C} \) \[1\]. This implies that the rate of NOₓ conversion can be accelerated by use of an oxidation catalyst upstream of the SCR unit, so as to convert ca. 50% of the NO to NO₂; this, in turn, enables the SCR catalyst volume to be reduced. Alternatively, the superior SCR kinetics obtained through use of an oxidation pre-catalyst provides an opportunity to reduce the operating temperature of the SCR system (at fixed SCR catalyst volume).

A specific objective of the project was to identify catalysts which are active for the oxidation of NO to NO₂ under typical flue-gas conditions, while possessing low activity for the oxidation of SO₂. Given that NO oxidation represents a key step in the functioning of NOₓ adsorber catalysts, commonly referred to as lean NOₓ traps (LNTs), the results of this project should also be relevance for the design of LNTs with reduced precious metal content.

A range of catalysts was screened for NO oxidation activity under conditions chosen to be representative of the flue gas from coal-fired utility boilers: \( T = 275-375 \, ^\circ\text{C}, \ [\text{NO}] = 250 \, \text{ppm}, \ [\text{SO}_2] = 0-2800 \, \text{ppm}, \ [\text{H}_2\text{O}] = 7\%, \ [\text{CO}_2] = 12\%, \ [\text{O}_2] = 3.5\%, \text{balance} = \text{N}_2 \). A number of catalysts were found to show excellent NO oxidation activity when SO₂ is absent from the feed gas, although in the presence of high SO₂ concentrations the conversion of NO is greatly suppressed (see figure). A detailed description of the results can be found elsewhere \[2,3\]

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