Alternative deNOx catalysts and technologies

The present thesis entitled Alternative deNOx Catalysts and technologies revolves around the topic of removal of nitrogen oxides. Nitrogen oxides, NOx, are unwanted byproducts formed during combustion (e.g. in engines or power plants). If emitted to the atmosphere, they are involved in the formation of acid rain and photochemical smog. Some basic concepts and reactions regarding the formation and removal of NOx are presented in chapter 1 and 2. Two approaches are undertaken in the present work to reduce the emission of NOx: by means of catalytic removal, and by NO absorption in ionic liquids. The commercial catalyst used for the selective catalytic reduction (SCR) of nitrogen oxides exhibits high activity and selectivity towards N2. However, the vanadia-titania-based catalyst used is very sensitive to deactivation by alkali-species (primarily potassium), which are typically present in high amounts in the flue gas when biomass is combusted. By co-firing with large amounts of CO2-neutral straw or wood (to meet stringent CO2 emission legislation), the lifetime of the traditional SCR catalyst is thus significantly reduced due to the presence of deactivating species originating from the fuel. To develop a catalyst less susceptible to the poisons present in the flue gas, a number of catalysts have been synthesized and tested in the present work, all based on commercially available supports. A highly acidic support consisting of sulfated zirconia was chosen based on preliminary studies. A number of different active species distributed on the support were investigated, such as iron, copper and vanadium oxides. However, based on the catalysts performance in the SCR reaction and their resistances towards potassium, the most promising candidate of the formulations studied was the vanadia-loaded catalyst, i.e. V2O5 - SO42- - ZrO2. This work, together with an introduction to the catalytic removal of NOx, are described in chapter 3. The remainder of the first part is concerned with the catalytic NOx removal (chapter 4) and it addresses the upscaling of the best catalyst candidate. The catalyst was mixed with the natural binding clay (sepiolite) to upscale the selected catalyst to the monolithic level, suitable for installation in gas stream with high flows, e.g. a flue gas duct of a power plant. A series of catalyst pellets with increasing levels of sepiolite were produced to evaluate the optimum mixing ratio. Based on these results, a monolith containing V2O5-SO2-4 -ZrO2 in 25 wt% sepiolite was produced, and evaluated with respect to the influence of space velocity, reaction temperature, and NH3/NO feed ratio on the NO reduction efficiency. The last part of this thesis deals with the non-catalytic sorption of NOx in ionic liquids, collected in chapter 5. Since no previous studies of the absorption of NO in ionic liquids have been reported, a preliminary study was conducted to identify suitable solvents. Two resulting imidazolium-based candidates, namely [BMIM]OAc and [BMIM]OTf, were selected due to their impressively high sorption capacities. Both solvents examined here revealed solubilities about twofold higher than those previously reported for e.g. CO2-capture in ionic liquids. Especially the [BMIM]OAc demonstrated extraordinary absorption capabilities, being able to retain around four NOmolecules per molecule ionic liquid. However, [BMIM]OTf exhibited promising behavior due to its reversible absorption/desorption properties. This in principle allows recycling of the ionic liquid as well as harvesting the NO. The accumulated NO could hereby be used in e.g. the synthesis of nitric acid allowing production of value-added chemicals from waste flue gas effluent. Although additional understanding of the mechanisms of the presented system is required, the perspective of a selective NO stripping technology is a very interesting alternative to the catalytic removal of NO from industrial flue gases.