

Ammonia Cracking: Catalysts and Process Configurations for Enhanced Performance

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Abstract : Compared to other hydrogen (H₂) carriers, ammonia (NH₃) is one of the most promising carriers as it contains 17.6 wt% hydrogen. It is easily liquefied at \approx 9-10 bar pressure at ambient temperature. More importantly, NH₃ is a carbon-free hydrogen carrier with no CO₂ emission at final decomposition. Ammonia has a well-defined regulatory framework and a good track record regarding safety concerns. Furthermore, the industry already has an existing transport infrastructure consisting of pipelines, tank trucks and shipping technology, as ammonia has been manufactured and distributed around the world for over a century. While NH₃ synthesis and transportation technological solutions are at hand, a missing link in the hydrogen delivery scheme from ammonia is an energy-lean and efficient technology for cracking ammonia into H₂ and N₂. The most explored option for ammonia decomposition is thermo-catalytic cracking which is, by itself, the most energy-efficient approach compared to other technologies, such as plasma and electrolysis, as it is the most energy-lean and robust option. The decomposition reaction is favoured only at high temperatures (> 300°C) and low pressures (1 bar) as the thermocatalytic ammonia cracking process is faced with thermodynamic limitations. At 350°C, the thermodynamic equilibrium at 1 bar pressure limits the conversion to 99%. Gaining additional conversion up to e.g. 99.9% necessitates heating to ca. 530°C. However, reaching thermodynamic equilibrium is infeasible as a sufficient driving force is needed, requiring even higher temperatures. Limiting the conversion below the equilibrium composition is a more economical option. Thermocatalytic ammonia cracking is documented in scientific literature. Among the investigated metal catalysts (Ru, Co, Ni, Fe, ...), ruthenium is known to be most active for ammonia decomposition with an onset of cracking activity around 350°C. For establishing > 99% conversion reaction, temperatures close to 600°C are required. Such high temperatures are likely to reduce the round-trip efficiency but also the catalyst lifetime because of the sintering of the supported metal phase. In this research, the first focus was on catalyst bed design, avoiding diffusion limitation. Experiments in our packed bed tubular reactor set-up showed that extragranular diffusion limitations occur at low concentrations of NH₃ when reaching high conversion, a phenomenon often overlooked in experimental work. A second focus was thermocatalyst development for ammonia cracking, avoiding the use of noble metals. To this aim, candidate metals and mixtures were deposited on a range of supports. Sintering resistance at high temperatures and the basicity of the support were found to be crucial catalyst properties. The catalytic activity was promoted by adding alkaline and alkaline earth metals. A third focus was studying the optimum process configuration by process simulations. A trade-off between conversion and favorable operational conditions (i.e. low pressure and high temperature) may lead to different process configurations, each with its own pros and cons. For example, high-pressure cracking would eliminate the need for post-compression but is detrimental for the thermodynamic equilibrium, leading to an optimum in cracking pressure in terms of energy cost.

Keywords : ammonia cracking, catalyst research, kinetics, process simulation, thermodynamic equilibrium

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