Getting $CuO/ZnO/ZrO_2$ in shape: Tableting of methanol catalysts

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Significance and Relevance

In this study a scientific approach towards catalyst shaping by tableting to enable a rational design of shaped catalyst bodies is presented. A moderate content of graphite as a lubricant was shown to be a compromise between a high mechanical stability and a sufficient level of lubrication, facilitating a continuous tableting process. Moreover, a variation of the compaction pressure revealed that the right choice could limit the drawbacks on desirable physico-chemical properties like porosity while keeping a high and even more reliable mechanical stability.

Preferred and 2nd choice for the topic: Preferred: CO_2 utilization and recycling, 2nd choice: Circular Economy

Preferred presentation: Poster

Introduction and Motivations

Shaping of catalysts into bodies, also called technical catalysts, is a major part of catalyst development in order to employ them in industrial reactors. Avoiding high pressure drops in fixed-bed reactors is one of the main reasons that make shaping of powdered catalysts necessary. While the catalytic performance of powdered catalysts is mainly the focus of academia, additional properties, such as mechanical stability and mass transfer, become important during the scale-up into technical catalysts.[1-3] Tableting is a well-established shaping method. Here, the catalyst is compacted between two punches into highly uniform and mechanically resistant bodies.[2,3] In general, catalyst shaping is a topic mainly driven by industrial manufacturers and in part governed by comprehensive empirical knowledge. Although of major importance for the performance and stability of the final catalyst, in-depth understanding of the processes at play and of their influence on the catalytic performance is still lacking in the open literature.[1,2] Therefore, systematic research of the shaping processes is important for a rational design of technical catalysts based on fundamental criteria and can help to bridge existing gaps between catalyst development in academia and industry.

Results and Discussion

Herein, a scientific approach towards catalyst shaping by tableting and its impact on the mechanical and physico-chemical properties is presented. For this purpose, a highly instrumented tableting machine was utilized, which is a powerful device for tablet development as it enables not only fundamental research of the compaction process and the resulting material properties, but also initial scale-up studies.

Tableting was studied for a co-precipitated $CuO/ZnO/ZrO_2$ catalyst for methanol synthesis from CO_2-rich synthesis gas.[4] At first, the effect of different parameters on the tableting process and the mechanical stability of the resulting flat cylindrical tablets was studied: pre-granulation procedure, content of the lubricant graphite, compaction pressure, compaction speed, tablet size as well as an additional pre-compaction step. Based on the results of the initial studies, a detailed investigation of the compaction pressure and the tablet density was enabled by a variation of the tablet weight for well-defined cylindrical mini tablets (2 x 2 mm), which were characterized by a broad range of measurement techniques.

The variation of the content of graphite used as a lubricant revealed a correlation with the ejection force of the tablets as well as the transmission ratio of the pressure from the lower to the upper punch. Lower graphite contents resulted a higher ejection force and a lower transmission ratio of the compaction pressure. Additionally, at the lowest graphite content the production of multiple tablets was accompanied by a continuous increase of the ejection force and decrease of the transmission ratio. Lubrication is improved at higher graphite contents leading to lower and constant ejection forces and higher transmission ratios. Here, a plateau is reached at moderate levels of graphite. In contrast, measurement of the side crushing strength showed that a higher mechanical stability is favored by lower graphite contents.

Based on these results, further detailed studies of compaction pressure and tablet density were conducted. For the mechanical stability a linear relationship was observed in the lower compaction pressure and tablet density range (Figure 1, left). In the higher range, the increase in crushing strength is less pronounced, less reproducible and accompanied by tablet defects. Porosity was studied by N_2 physisorption and Hg porosimetry to link the changing contributions of micro-, meso- and macropores to the overall porosity with the compaction pressure (Figure 1, right). Tableting was shown to have a negative impact on the reducibility, which was evaluated by temperature-programmed reduction. Increasing compaction pressures led to higher reduction temperatures, a broadening of the reduction peaks and lower hydrogen uptake. Moreover, reduced tablets were smaller in size and showed a change in mechanical resistance, depending on their initial density.

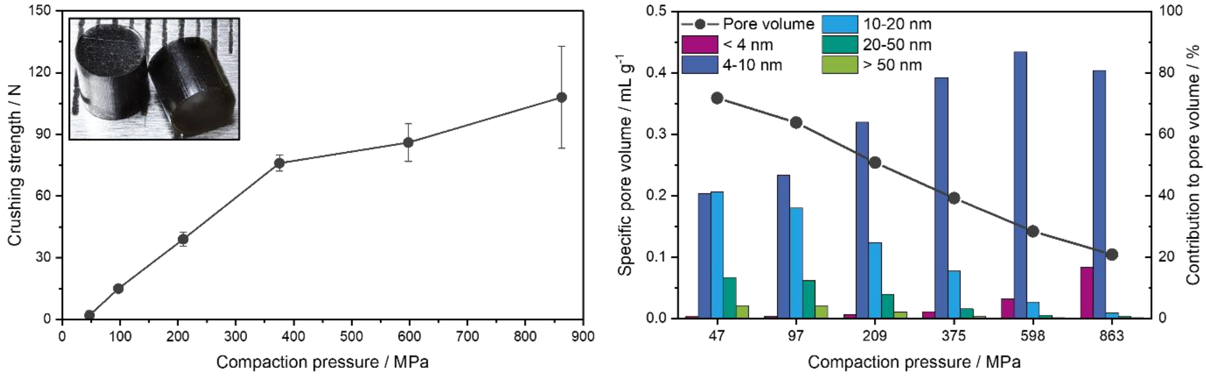

Figure 1: Crushing strength (left) and specific pore volume and contribution of different pore size ranges to the pore volume (right) of cylindrical tablets (2 x 2 mm) in dependence of compaction pressure.

The presented study shows that a moderate graphite content is necessary for a high mechanical stability, but also to have sufficient lubrication enabling continuous tablet production. Increasing the compaction pressure and tablet density exemplified the expected trade-off between mechanical stability and desirable physico-chemical properties like porosity. The results however show that with less than 50% of the highest compaction pressure investigated, similar and more reliable mechanical stabilities can be achieved while avoiding further negative effects on porosity and reducibility.

References

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