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Process Improvements by the Utilisation of an Annular Slit Reactor

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Abstract. For a strongly endothermic catalytic dehydrocyclisation a new design of an annular slit reactor was evaluated. The major advantage of the design is an optimised temperature profile. Consequently, both a higher selectivity and a higher conversion were achieved. Furthermore, the formation of a problematic side product was significantly reduced, leading to a simplification of the subsequent purification step and an increased productivity. From an economic point of view, yearly savings of up to CHF 1 Mio. are anticipated compared to the present manufacturing costs.

Introduction

Presently, the manufacturing of a multistep pharmaceutical active substance at *Lonza AG* involves the performance of a catalytic dehydrocyclisation at a temperature above 600°. The strongly endothermic reaction is characterised by the formation of the desired product with simultaneous liberation of hydrogen.

In order to accomplish such heterogeneous gas-phase reactions on an industrial scale, various types of reactors have been proposed in the literature. In *Lonza*'s case a readily available tubular reactor with an external electrical heating system was selected. This choice was based on an immediate need of a manufacturing capability rather than on a detailed technical and economical consideration.

The simplicity of the construction and an easy loading and unloading of catalyst can be seen as advantages of such a fixed-bed reactor. On the other hand, the occurrence of distinctly different temperature profiles in radial and axial directions must be expected with reactions characterised by large enthalpy changes. This disadvantage is clearly discernable in the tubular reactors set up for the industrial production at *Lonza*. The construction characteristics of the reactors (l = 3 m, d_i = 0.1 m) in combination with a poor heat transfer in the catalytic bed result in unacceptable temperature profiles in both the radial and axial directions, *e.g.* temperature differences of up to 50° are observed in radial directions.

As a consequence, both concurrent and consecutive side reactions are favoured and result in modest selectivity and yields for the desired product. Furthermore, the partial depositing of side products causes a continuous decrease in the catalyst activity, requiring a regeneration by steam washing once a day. The major drawback of the modest selectivity is highlighted in the subsequent purification of the crude product. The reduction of the critical side product to the specified limit requires an additional costly rectification and ultimately becomes the bottleneck of the process.

Annular Slit Reactor

Continued development work in the catalyst group of *Lonza AG* clearly demonstrated that by enhanced controllability of the temperature profile significant improvements can be achieved in respect to selectivity, yield, and catalyst 'life-time'.

To attain both the necessary reaction-temperature profile and the requirements of high specific throughput and low pressure drop due to the release of hydrogen, an annular slit reactor was proposed (*Fig. 1*). Such a reactor consists of two concen-

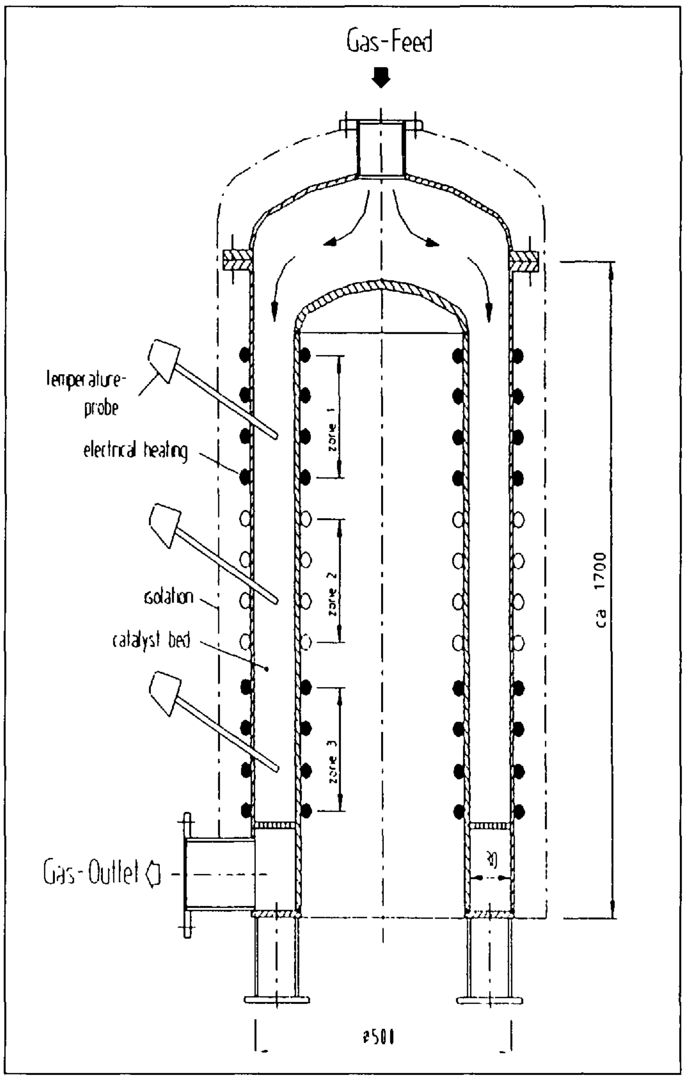

Fig. 1. *Scheme of an annular slit reactor*

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tric tubes, in which the annular space defined by the inner and outer tube is packed with the catalyst. The advantage of such a design lies in the possibility of reducing the thickness of the catalyst bed by one third without losing on the specific throughput.

In order to further improve the specific heat input in radial directions, an electrical heating system is installed both on the inner and outer tube wall. The heating of the catalytic bed in axial direction is divided in three zones. Each zone has its independent electrical heating system. By this manner, the strongly changing heat demand due to the endothermic reaction can be better satisfied. Good controllability, relatively easy construction, and safety considerations gave this heating system the preference compared to one heated by means of molten metal or gas.

Piloting Results

A pilot reactor according to *Fig. 1* was set up for the further development of the process. Equal specific throughput and cycle time of 23 h (20 h reaction, 3 h regeneration) were applied for the piloting as it is presently practised in the industrial tubular reactors.

To explore the parameter sensitivity and to identify the optimal temperature profile, a statistical experimental design was applied. After a first parameter screening was successfully performed, the fractional factorial design was enlarged to a central-composite design. As a result, a mathematical model was generated for the three most important parameters, these being the temperatures in each of the heating zone in axial direction. With the aid of this quadratic model, it becomes feasible to predetermine the process conditions for highest selectivity and yield, or maximum yield at a given side-product level.

A comparison of typical yield and side-product level towards the end of the reaction cycle shows the superior performance of the annular slit reactor compared to the conventional tubular reactor (*Table*). The corresponding temperature profile in axial direction is presented in *Fig. 2*. In radial direction a temperature difference of < 15° can be achieved with these conditions.

As outlined in the introduction section, productivity is limited by the presence of a side product, which has to be partially removed by subsequent rectification. As a consequence, the correlation between this crucial side-product formation in relation to the expected yield was identified with the statistical model (*Fig. 3*). Additionally, the optimisation algorithm also identified the appropriate operating conditions, which were than applied to the pilot reactor. As can be seen, verification of the model is within experimental error (*Fig. 3*).

Table. *Comparison of Typical Yield and Side-Product Levels after 1 and 18 h of Production*

Typical performance drop between 1 and 18 h after restart	Selectivity [%]	Yield [%]	Content of critical side product [%]
tubular reactor	65 → 57	52.4 → 39.8	5.1 → 4.0
annular slit reactor	81 → 78	61.8 → 56.7	3.6 → 3.0

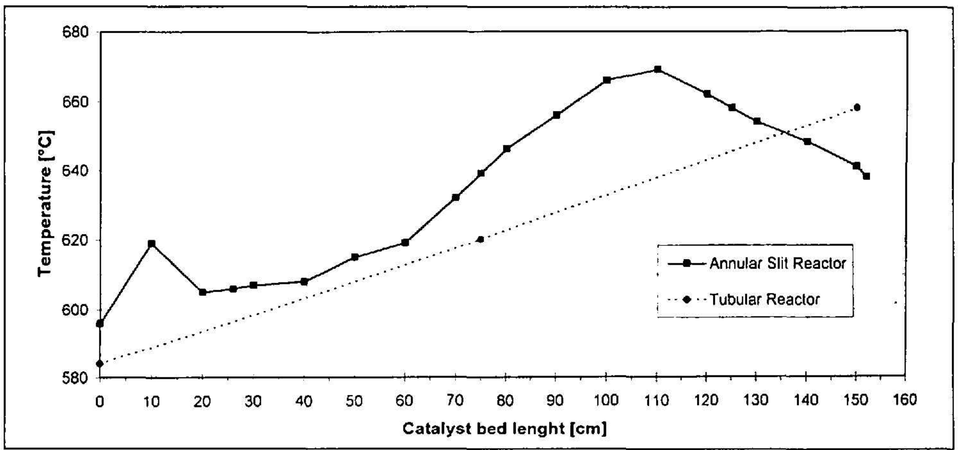

Fig. 2. *Comparison of the temperature profiles*

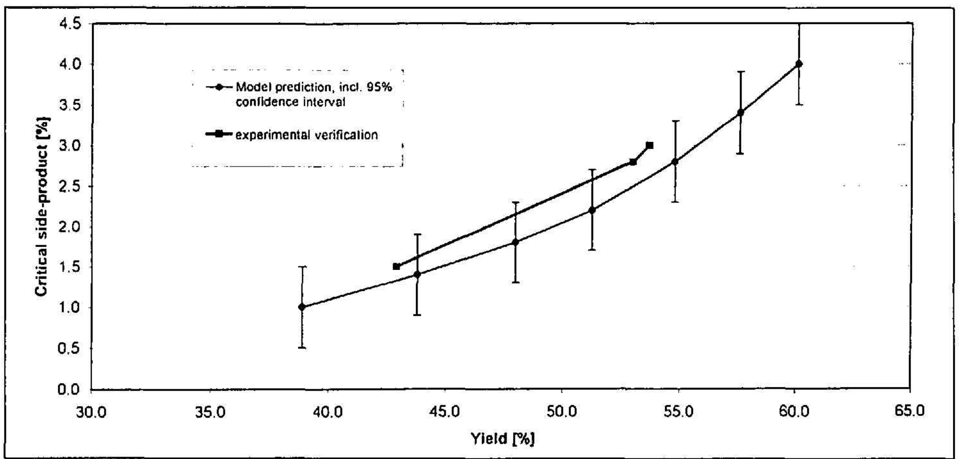

Fig. 3. *Correlation between critical side-product and expected yield*

Concluding Remarks

The piloting results have clearly demonstrated the advantages of an annular slit reactor (*Fig. 1*) in comparison to a conventional tubular reactor for strongly endothermic reactions at elevated temperatures (*Table*). Only the enhanced specific heat input combined with an improved controllability of the heating system makes the adaptation of the optimal temperature profile within the catalytic bed feasible. In particular, it enables the optimisation of a crucial side-product formation in respect to quality and productivity for the subsequent purification step. Furthermore, reduced side-product formations due to higher selectivity will improve the catalyst 'life-time' and ultimately result in an extended production cycle time.

Presently, ten tubular reactors are in operation at *Lonza AG* for the industrial performance of the described gas-phase reaction. However, these conventional reactors will be replaced by two annular slit reactors in the near future. Several process improvements, based on experiments described in this paper, are thus expected on industrial scale. From an economical point of view, yearly savings of up to CHF 1 Mio. are anticipated compared to the present manufacturing costs.

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