

## Distillation column with reactive pump arounds: an alternative to reactive distillation

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### Abstract

The hardware design of reactive distillation (RD) columns pose severe challenges with respect to the choice and design of the hardware; the requirements of reaction (i.e. high liquid or catalyst holdup) is not in consonance with the requirement of separation (high interfacial area). In this paper, we examine an alternative to the RD concept, namely a distillation column networked with a number of side (external) reactors. If each distillation stage is linked to a side reactor, the performance of the RD column is matched exactly. From a practical point of view, it is desirable to reduce the number of side reactors to below, say, six. The precise location of the chosen number of side reactors and the manner in which the liquid draw-offs and reactor effluent re-entry to the distillation column needs to be chosen carefully. We have developed an algorithm to determine an optimum configuration of the side-reactor concept in order to maximise conversion. For the case study of methyl acetate production, we see that it is possible to match the conversion level of an RD column by appropriate choice of the number of side reactors and the pump around ratio. The higher the conversion target the larger the number of side reactors and pump around ratios. For modest conversion levels, say <90%, even a 3-side-reactor configuration will be able to match the performance of the RD column. The study presented here reveals the potential, and limitations, of the side-reactor concept for use as an alternative to RD technology.

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### 1. Introduction

Reactive distillation (RD) is enjoying a lot of attention from industry and academia because of the many advantages over the conventional reaction followed-by separation concept [1–4]. The successful commercialisation of RD technology requires special attention to hardware design that does not correspond to those for conventional (non-reactive) distillation. In most RD applications the Hatta number is less than unity [5] and therefore, there is no enhancement of mass transfer due to chemical reaction. In order to maximise productivity in a homogeneously catalysed RD column, we need to maximise the liquid holdup. In a heterogeneously catalysed RD column, we similarly need to maximise the

catalyst holdup. The requirements of high liquid or catalyst holdup in an RD column are not in consonance with the requirement of good in situ separation, for which we need to maximise the interfacial area between vapour and liquid. All available hardware configurations (tray or packed RD columns) represent a compromise between the conflicting requirements of reaction and separation [3,4].

There is another issue that mitigates the idea of carrying out the reaction within a distillation column; this relates to catalyst deactivation. The traditional way to compensate for the catalyst deactivation, i.e. adding excess catalyst or increasing the reaction temperature, is seldom feasible in RD applications.

One way to overcome the above mentioned hardware problems with RD columns, while maintaining the benefits of in situ separation with reaction, is to employ the side-reactor or external reactor concept [6,7]; see Fig. 1. In the side-reactor concept the reactor feed is withdrawn from the distillation column and the reactor effluent is returned back to the same column. The side reactor could be a conventional catalytic-packed bed reactor operating in liquid

*Abbreviations:* HOAc, acetic acid; MeOAc, methylacetate; MeOH, methanol

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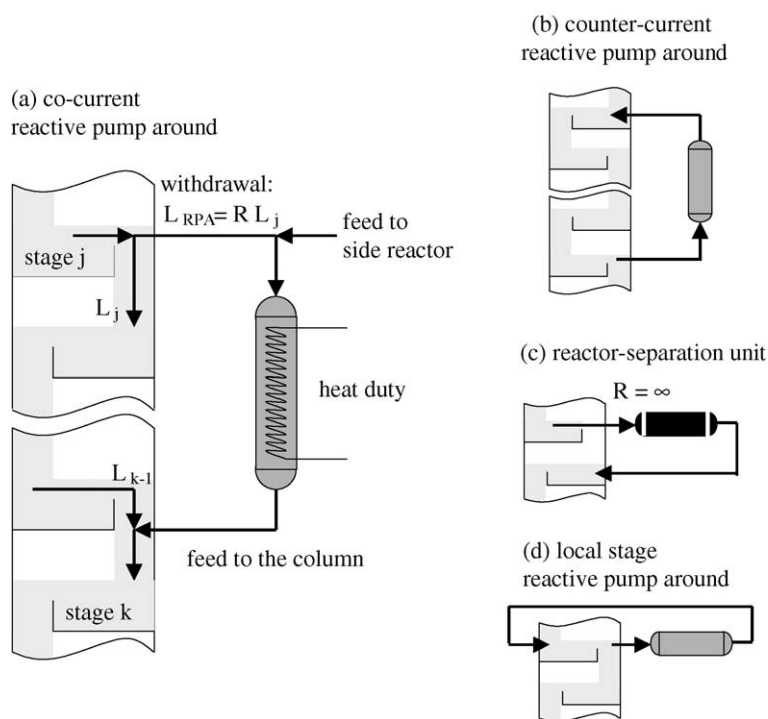


Fig. 1. Schematic diagram of side-reactor configurations.

phase and therefore, there are no hardware design problems or conflicting requirements with regard in respect of in situ separation. Furthermore, the reaction conditions within the side reactor (e.g. temperature) can be adjusted independently of those prevailing in the distillation column by appropriate heat exchange.

In principle, we can distinguish four configurations for linking the side reactors to the distillation column; these are shown in Fig. 1(a)–(d). The pump around can be located in such a way that liquid bypasses intermediate stages (see Fig. 1(a)). The liquid is withdrawn from stage  $j$  and possibly mixed with an additional feed stream before it is pumped to a side reactor. The stream leaving the side reactor is fed back to the column at stage  $k$ . The amount of liquid pumped-around,  $L_{RPA}$ , can either be specified by an absolute molar flow rate or by a ratio,  $R$ , with regard to the molar flow entering the stage below,  $L_j$ . In this case the reactor throughput is limited to a maximum fraction of the internal flows in the distillation column. Increasing the pump around flow rate above that limit would dry out the intermediate stages in the column, and significantly decrease the separation performance. We call this configuration *co-current reactive pump around*. Fig. 1(b) shows an alternative configuration where the side-reactor flows are *counter-current* to the internal liquid stream in the distillation column. The throughput in the reactor can exceed the original internal flows in the distillation column, but also might raise the danger of flooding on intermediate stages and demands additional energy input. Both configurations, (a) and (b), cause a change of internal flow rate, which affects the operation line. This can cause the

driving force for mass transfer to decrease or energy losses to increase.

Co- and counter-current reactive pump around configurations have two limiting cases shown in Fig. 1(c) and (d), respectively. The product stream of side reactor might be fed to the downcomer at the same stage (see Fig. 1(d)). In this case, same limitations apply as already mentioned for a counter-current configuration. Of practical importance is the case where the stream leaving a stage is completely re-routed through a reactor before it is fed back to the stage below (see Fig. 1(c)). We call this configuration a *reactor-separation unit*. Since no stages are bypassed, the entire liquid stream leaving the stage will be pumped through the reactor. If the catalyst load is small enough to be placed in the downcomer an external reactor is not required [3].

In order to meet the process requirements of conversion, more than one side reactor may be required. Clearly, the determination of the optimum number of side reactors, along with the liquid draw-off and feed-back points to the distillation column need careful attention and consideration. The first major objective is to compare the performance of the side-reactor concept with a conventional RD column with regard to the liquid, or catalyst holdup. The second major objective of the present communication is to develop an algorithm to determine an optimum way to connect a given number of side reactors to a distillation column in order to maximise the conversion. We demonstrate our algorithm by considering the case study for production of methyl acetate.

In the modelling to be presented below, the reactor itself is modelled by a series of single phase CSTR reac-

tors, where the catalyst load and heat duty is evenly distributed. By choosing a sufficient number of CSTR reactors in series, say  $N_{CSTR} = 10$ , the reactor represents an ideal plug flow reactor. The model also allows operating the reactor adiabatically, isothermally, or with external heat supply.

## 2. Case study of methyl acetate synthesis

Consider the production of methyl acetate (MeOAc) by the acid-catalysed esterification reaction of acetic acid (AcOH) with methanol (MeOH). This reaction is made difficult by a variety of factors: (a) reaction equilibrium limitations, (b) difficulty of separating AcOH and H<sub>2</sub>O and (c) presence of MeOAc–H<sub>2</sub>O and MeOAc–MeOH azeotropes. Conventional processes use one or more liquid-phase reactors with large excess of one reactant in order to achieve high conversions of the other. In the conventional process the reaction section is followed by eight distillation columns, one liquid–liquid extractor and a decanter. This process requires a large capital investment, high energy costs and a large inventory of solvents. In the RD process for methyl acetate, invented by Eastman Chemical Company [1,8], the process is carried out in a single column. In

this single column, high-purity methyl acetate is made with no additional purification steps and with no unconverted reactant streams to be recovered. By flashing off the methyl acetate from the reaction mixture, conversion is increased without using excess of one of the reactants. The reactive column has stoichiometrically balanced feeds and is designed so that the lighter reactant MeOH is fed at the bottom section and the heavier acetic acid is fed at the top. The column consists of three sections. The reaction takes place predominantly in the middle section. The bottom section, serves to strip off the MeOH from water and return it to the reaction zone. The vapours leaving the reactive section consists of the MeOAc–MeOH azeotrope which is “broken” in the rectifying section by addition of AcOH which acts as an entrainer.

The starting point for our studies is the conceptual design of the RD column discussed in detail by Doherty and Malone [1]. The column operates at a pressure of 0.1 MPa. We employed a partial reboiler and a total condenser. The non-reactive section contains 10 theoretical stages and 33 catalytically active theoretical stages in the reactive section. The reactants methanol and acetic acid are fed stoichiometrically with a feed flow rate of 280 kmol/h on the Stages 5 and 40 (see Fig. 2(a)). A pseudo-homogenous rate model adopted from Doherty and Malone [1] describes the

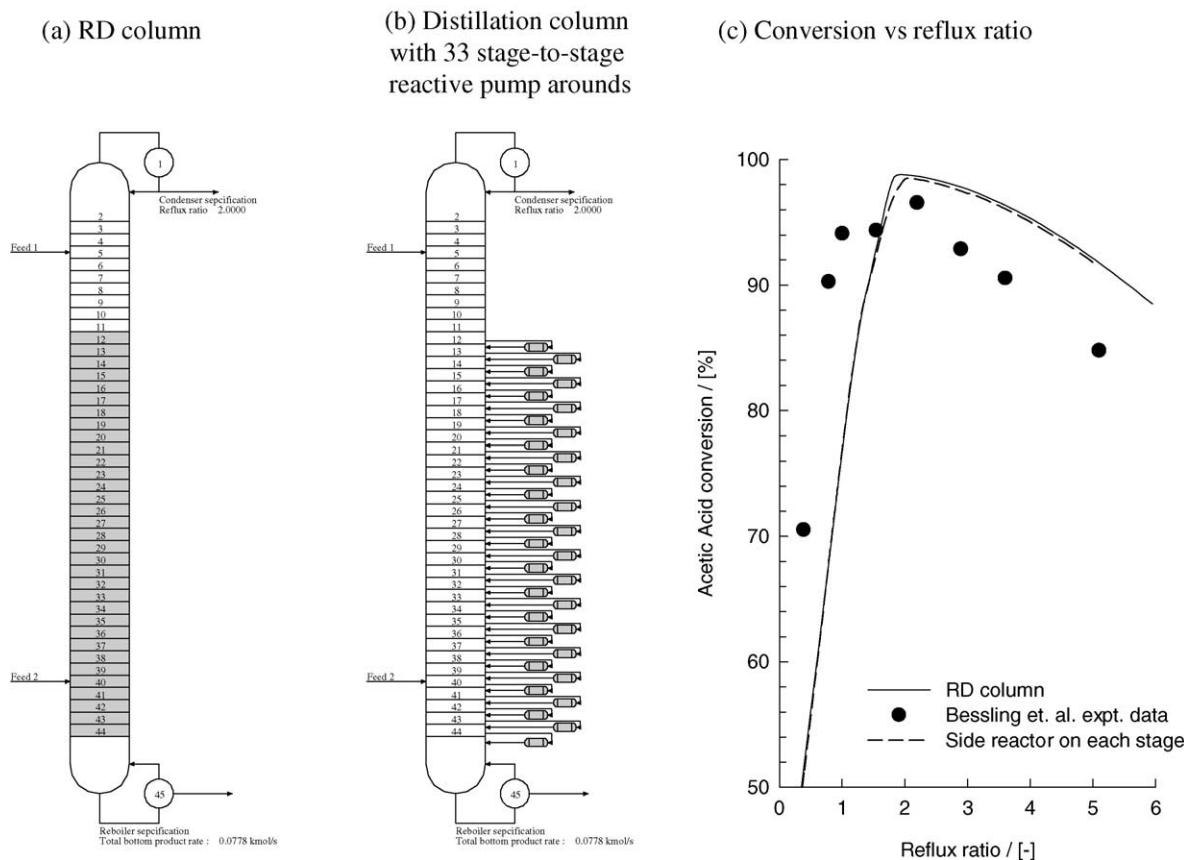


Fig. 2. Comparison of HOAc conversion between a reactive distillation column and a column designed by means of a reactor–separation concept when the reflux ratio is varied. The overall catalyst load is 99 m<sup>3</sup> in both cases.

reaction. The rate expression is given by:

$$r = c_1 k_f \left( a_{\text{HOAc}} a_{\text{MeOH}} - \frac{1}{K_{\text{eq}}} a_{\text{H}_2\text{O}} a_{\text{MeOAc}} \right) \quad (1)$$

with the forward reaction rate constant:

$$k_f (\text{s}^{-1}) = 2.7033 \times 10^5 \left( \frac{-6287.7}{T} \right) \quad (2)$$

and the equilibrium constant:

$$K_{\text{eq}} = 2.32 \exp \left( \frac{782.98}{T} \right) \quad (3)$$

The liquid phase activity coefficients are very well represented with Wilson parameters, reported by Doherty and Malone [1] who also give the vapour pressure data. We did not consider the formation of the side product dimethyl ether and water from methanol. Furthermore, the side reaction has a minor impact on the column performance at atmospheric pressure [1]. A macro-reticular ion-exchange resin such as Amberlyst 15W is used as a catalyst. The catalyst is introduced at the top of the reactive section. Further, we assumed that the volumetric liquid holdup on a theoretical stage is  $3 \text{ m}^3$ . We first consider a distillation column with 33 stage-to-stage reactive pump-arounds; see Fig. 2(b). The bottom product flow rate is fixed at  $280 \text{ kmol/h}$ , whereas the reflux ratio is varied. The catalyst load in each of the 33 reactors corresponds to  $3 \text{ m}^3$  catalyst load on each reactive stage in the reactive distillation column. Calcula-

tions of the conversion in the RD column (Fig. 2(a)) and the 33-side-reactor configuration (Fig. 2(b)) using the EQ stage model [3,9] are shown in Fig. 2(c) for varying reflux ratios. The results are in reasonably good agreement with the experimental results of Bessling et al. [10]. As can be seen from the Fig. 2, the separation–reaction unit concept is equivalent to an RD column. In cases where the catalyst load is small, the catalyst can be placed in the downcomer and so functions as a reactor [9]. If the space in the downcomer were not sufficient, one would have to connect 33 reactors to the column. This is practically not a desirable design and would result in high investment costs. Therefore, our objective is to reduce the number of reactors and find configurations with high selectivity and conversion.

### 3. Search algorithm for locating reactive pump arounds

In order to locate and determine the side reactors, we developed an algorithm based on a simple one-dimensional search; see Fig. 3. Considering the fact that catalyst load, heat duty, flow rate and locations of the pump arounds are unknown parameters, it is advisable to reduce the complexity of the problem in the first step. Therefore, we assume that the side reactor operates adiabatically and at chemical equilibrium. We also fix the operating conditions for the

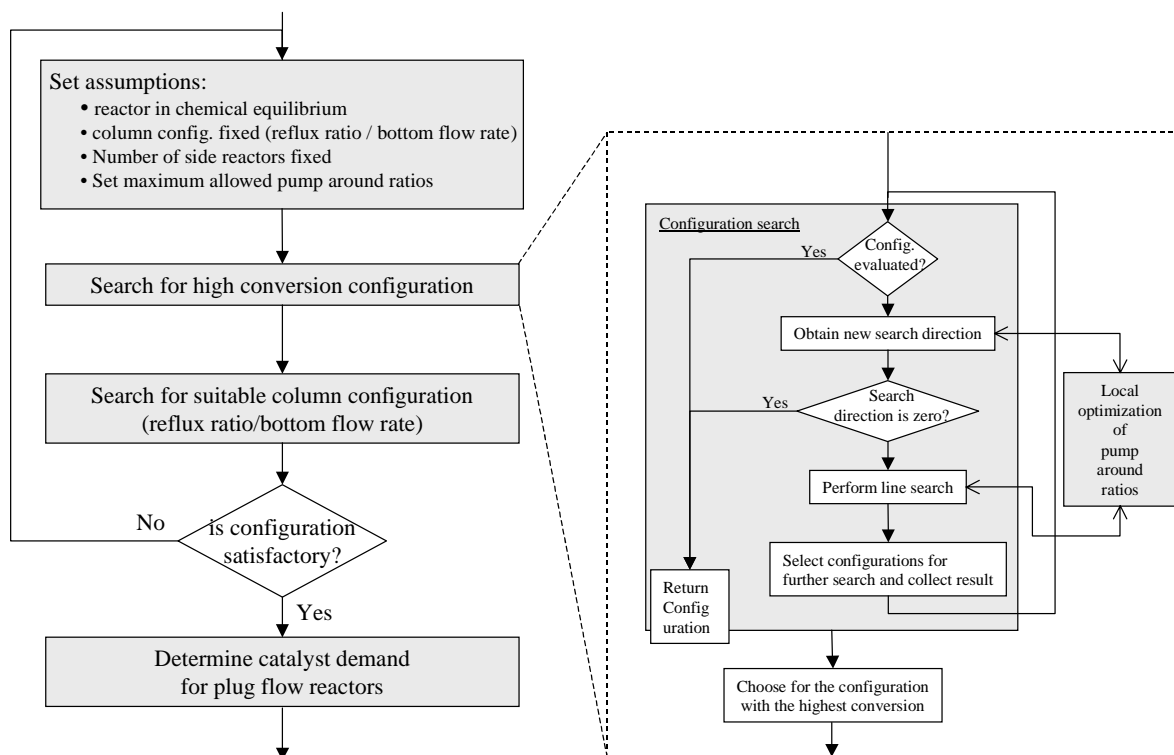


Fig. 3. Simplified diagram of the search algorithm used to determine side-reactor configurations.

distillation column. Once the algorithm has placed the side reactors, we will check whether the column performance is satisfactory. If this is the case, we accept the configuration. If not, it is advisable to restart the search with a modified operation point.

The search algorithm has to fix the location and throughput of  $N_{\text{RPA}}$  reactive pump arounds. Hence, we have to determine  $N_{\text{RPA}}$  continuous parameters for the pump around throughputs and  $2N_{\text{RPA}}$  discrete parameters for the in- and outlet locations of the side reactors. The pump around ratio with regard to the liquid flow leaving the stage above determines the throughput of a side reactor. Simulations showed that evaluating a configuration with slightly different pump around locations might already be hard to converge due to significant changes of internal composition and flow profiles. In particular, this is true if the pump around ratio exceed unity. Therefore, we decided to employ a line, i.e. one-dimensional, search algorithm with a limiting step size of moving an in- or outlet of a side reactor by only a single stage up or down. The search direction for the discrete line search algorithm is determined by the steepest conversion increase when the locations of single side reactors are varied. This implies that the  $N_{\text{RPA}}$  reactive pump arounds are decoupled. Hence, in order to limit computational costs not all-possible search directions are considered. We follow the search direction until a continuing decrease of conversion is detected for a consecutive number of iterations. This is done in order to pass small local maxima caused by small local variation in conversion when following the search direction. The local maxima along the search path are recursively evaluated. Finally, the algorithm terminates if no search direction is found in which conversion increases by locally relocating a single reactive pump around. It should also be clear that with the line search algorithm for location of the side reactors, the optimum solution could be one of the four types shown in Fig. 1.

Each time a new configuration is evaluated, an inner optimisation using a conjugate gradient approach calculates the optimal throughput. In most cases, increasing throughput, i.e. increasing pump around ratios, results in an increase of conversion. Since our objective function is restricted to the overall conversion, it is useful to introduce a limiting value,  $R_{\text{max}}$ , for the pump around ratio. In case of a reactor–separation unit (Fig. 1(b)), the limiting value is always set to  $R_{\text{max}} = 15$ , which corresponds to a throughput of over 90% of the flow leaving the stage under consideration.

The algorithm described above is straightforward and pragmatic. Several simulations under equal conditions but with different initial configurations resulted in similar and comparable final configurations. Although the algorithm does not necessarily detect local optima, it provides useful information about promising configurations that serve for further investigation. The resulting configurations also give a good starting point for detailed configuration refinements.

#### 4. Side-reactor configurations

On the basis of the information in Fig. 2(c), we choose a reflux ratio of two in the optimisation studies for side reactors. In order to study the influence of the throughput through each side reactor we varied the limits of  $R_{\text{max}}$ , for co- and counter-current reactive pump arounds, whereas for a reactor separation unit,  $R_{\text{max}}$  is always chosen to be 15. We determined optimal configurations for columns with 1–6 side reactors. For every choice of the number of side reactors, we consider the choice of the pump around ratio  $R_{\text{max}} = 0.1, 1-3, 5$  and 10. Fig. 4 shows a selection of column configurations obtained with two, three and five side reactors. Typical production rate and MeOAc composition profiles are depicted in Fig. 5.

A careful examination of all our simulation results reveals some common observations with regard to the proposed side-reactor configurations:

- High reactor throughput due to high pump around ratios and counter-current configurations are beneficial for conversion.

Counter-current operating reactive pump around causes an increase of the internal liquid flows and therefore, results in larger reactor throughput. Further, the inner optimisation loop for the pump around ratio almost always hits its maximum limit  $R_{\text{max}}$  in order to provide a large throughput. Major drawback of high pump around ratios and counter-current operation is the high energy demand.

- When pump around ratios are restricted to comparable low pump around ratios, a reactor–separation unit gets attractive. However, bypassing stages, i.e. co-current reactive pump around configurations, were not found to be beneficial in the present case study.

Co-current reactive pump around were not found to be beneficial since (a) the reactor throughput of a co-current reactive pump around is limited to a fraction of the internal flows, and (b) low internal flow rates on the intermediate stages cause a decrease in separation. In case of a reactor separation unit the throughput is nearly equal to the liquid flow leaving the column and no stages are bypassed. It turned out that for low pump around ratio such a configuration could be superior to a counter-current configuration.

- Recycle flows caused by counter-current side reactor configurations significantly influence the thermodynamic driving forces on intermediate stages. As a consequence it is preferable to reduce the number of intermediate stages when the recycle flow rates are large.

Mixing effects of the reactor flow and the internal column flow have a significant impact on the separation capabilities of intermediate stage in a counter-current side reactor configuration. Therefore, the thermodynamic driving forces for mass transfer and the reactor inlet composition of the side reactor, i.e. the chemical driving force in the side reactor, are affected. For the limiting case of

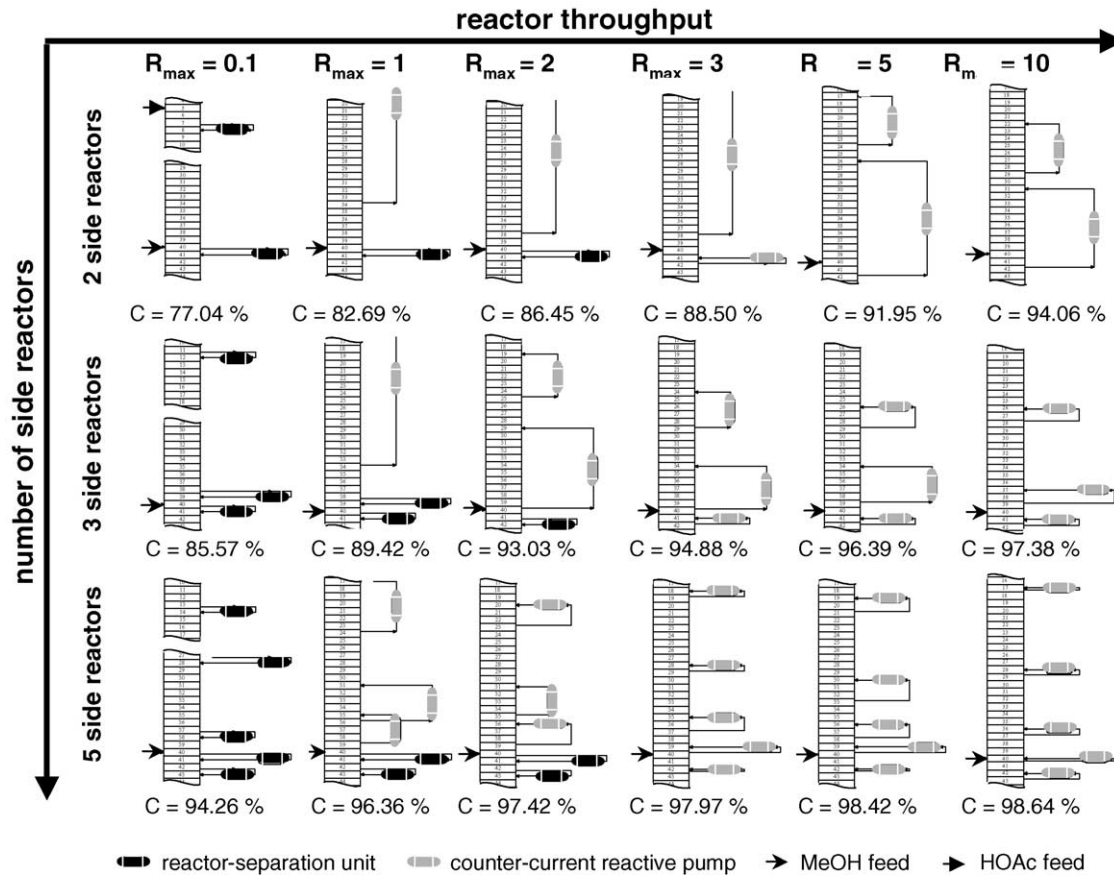


Fig. 4. Column configurations with two, three and five side reactors.  $C$  denotes the overall HOAc conversion and  $R_{max}$  the maximum limit of the reactive pump around ratio.

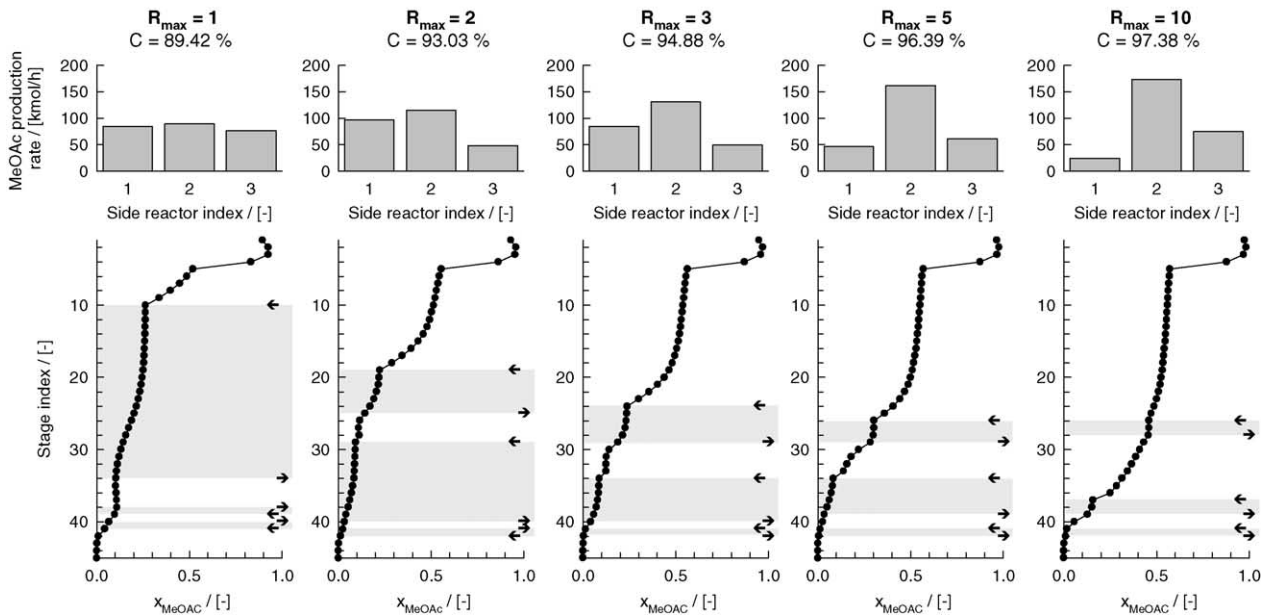


Fig. 5. Column configurations with three side reactors.  $C$  denotes the overall HOAc conversion and  $R_{max}$  the maximum limit of the reactive pump around ratio. The production rates and the MeOAc composition profile along the column height are shown for each configuration. The grey shaded area in the MeOAc composition profile denotes a region with changed internal flow rates due to reactive pump arounds. The arrows pointing towards the y-axis denote the outlet location of a side reactor and the arrows pointing away from y-axis denote the inlet location of a side reactor.

very high pump around ratios the concentration gradient between the inlet and outlet of the side reactor nearly vanishes. Hence, assuming the side reactor to operate at chemical equilibrium, all liquid compositions on the intermediate stages are forced close to chemical equilibrium. This is not advantageous for separation and so the number of intermediate stages is obviously minimised, for very high pump around ratios the recycle could even involve only a single stage. Figs. 4 and 5 also indicates that with increasing recycle, i.e. increasing pump around ratios  $R_{max}$ , fewer intermediate stages are preferred.

- Large recycle flows cause small chemical driving forces in the side reactors. This will result in high catalyst demand.

The chemical driving force of the side reactor also vanishes with fading composition gradient between the reactors in and outlet. This effect, however, is compensated by a large throughput that maintains high conversion. In practice such configurations with small chemical driving forces will, however, require large catalyst loads.

Fig. 6 summarises the performance of all configurations with regard to conversion versus pump around ratio. The reflux ratio is two and the bottom flow rate is 280 kmol/h. As expected, additional side reactors and high reactor throughput lead to improved performance. The limiting cases for small pump-around ratios are column configurations with solely reactor–separation units. It is an economical trade off between configurations with larger recycle flows and, in return, with fewer side reactors or with lower throughput but more side reactors. The first choice calls for high catalyst loads and energy costs, whereas latter one results in higher investment costs.

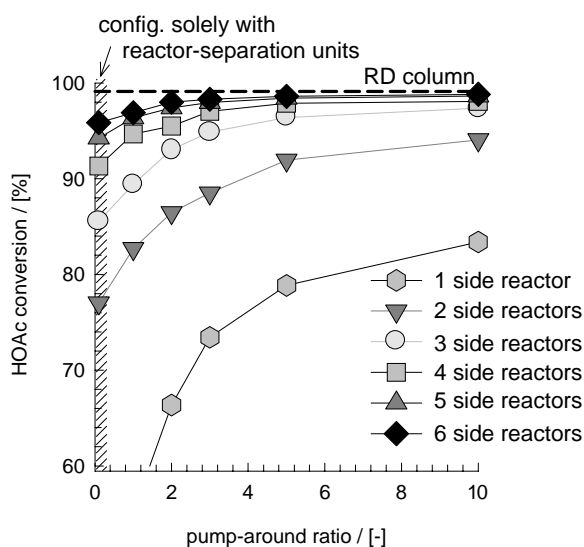


Fig. 6. Diagram summarising HOAc conversion for the configurations with 2–6 side reactors. The reflux ratio is two and the bottom flow rate 280 kmol/h.

## 5. Operation considerations

Fig. 7 shows the homotopy diagram of MeOAc conversion with respect to reflux ratio and bottom flow rate for columns with three and five side reactors. Also shown in Fig. 7 is the performance of the RD column. Except for configuration consisting solely of reactor–separation units, the chosen operation points for columns with side reactors are satisfactory. Furthermore, we observe a qualitative match with the behaviour of a comparable RD column. This indicates that in case of the present study the design guidelines for reactive distillation appear to be valuable tools when designing a column with side reactors.

- In order to increase the throughputs of reactor–separation units, it is beneficial to operate with higher internal flow rates. Hence, it can be favourable to increase reboil-ratio, heat duty or reflux ratio.

Fig. 7 shows that configurations with solely reactor–separation units require higher reflux ratios for maximum conversion. Increasing the reflux ratio results in an increase of the internal flow rates and so leads to larger reactor throughputs. This is beneficial for conversion. For three reactor–separation units, we can observe that the configuration is even superior to a combination of counter-current reactive pump-arounds and reaction–separation units; see Fig. 7(a). Increasing the internal flow rate, however, has the drawback of larger column diameters and higher energy costs. Hence, it might be more economical to operate a column with counter-current side reactors at higher pump around ratios and so partially higher liquid but smaller vapour flow rates.

Previously we had stated that side-reactor configurations qualitatively match an RD column. This is not true in all cases. Particularly, a mismatch can be expected if the RD column exhibits multiple steady states. Doherty and Malone [1] also reported the existence of multiple steady states for the MeOAc production. Subsequently, we illustrate that the steady state topology significantly depends on the side-reactor configurations. Therefore, we computed HOAc conversion of a column with two and five side reactors for a wider range of operation points, i.e. for reflux ratios up to 10 and bottom flow rate varying from 260 to 300 kmol/h. The column configurations correspond to the ones in Fig. 4 and were designed for a reflux ratio of two and a bottom flow rate of 280 kmol/h. As can be seen, multiple steady states are not found in the vicinity of the design operation point (see also Fig. 7). Above a reflux ratio of two, the RD column exhibits only a narrow region with three steady states. In case of columns with side reactors, we observe much wider regions with three or five multiple steady states; see hatched areas in Fig. 8. In some cases we even were able to identify very narrow regions with seven steady states. The complicated steady state topology for side-reactor configurations might be attributed to the feedback effects. However, further investigations, which are beyond the scope of our study, are needed to identify the exact causes and mechanisms.

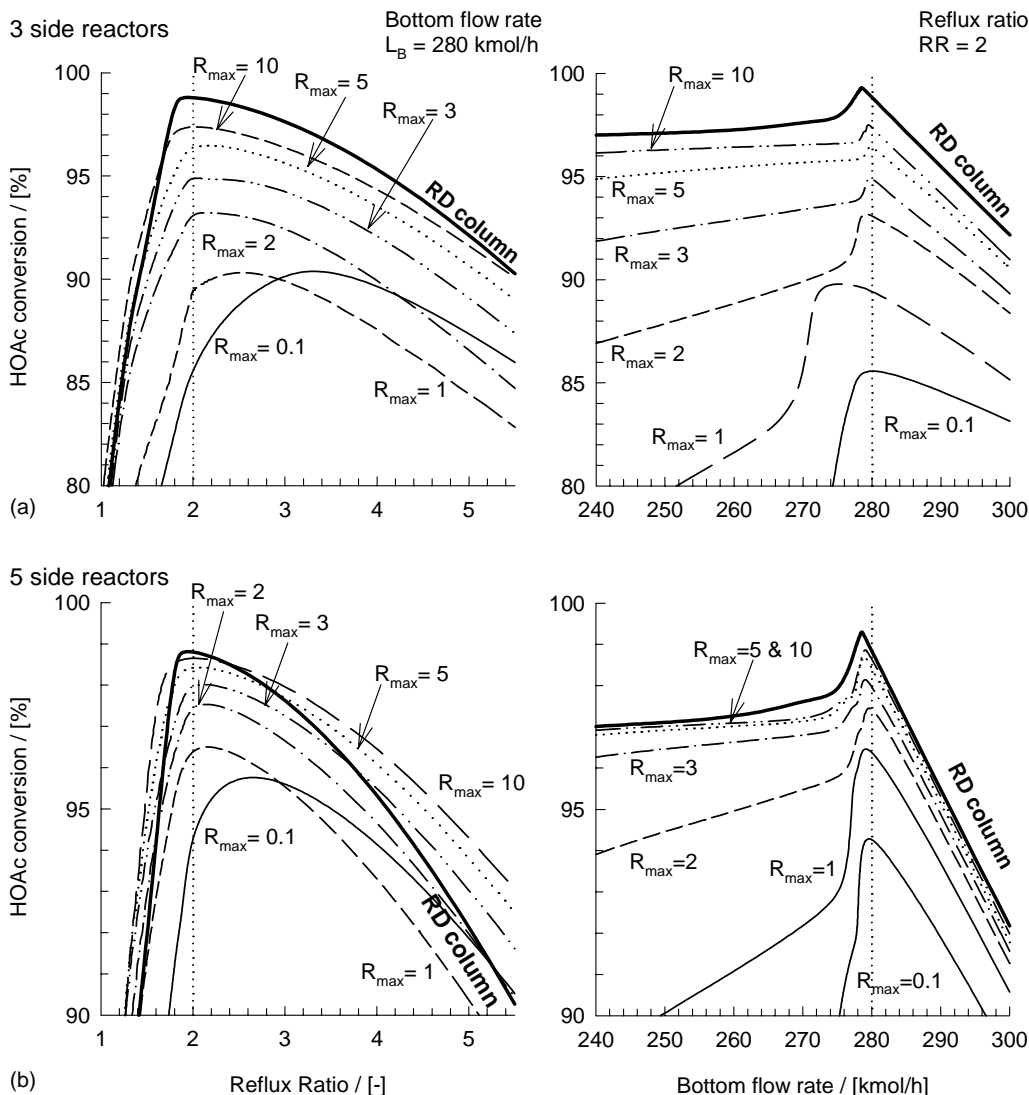


Fig. 7. Homotopy diagrams of HOAc conversion with regard to reflux ratio and bottom flow rate for configurations with (a) three and (b) five side reactors.

One should keep in mind that the steady state topology differs for each configuration and can be significantly more complex than the one for an RD column. This will also reflect on the dynamic behaviour and might require a sophisticated control design. Furthermore, when designing a side-reactor configuration, it is important to be aware of multiple steady states (if existent). Otherwise there will be the danger that the objective function of an optimisation or search algorithm is misleading.

## 6. Side-reactor feed

Fig. 9(a) shows configurations for columns with two side reactors where MeOH is directly fed to one of the side reactors. Comparing the conversion levels in Fig. 9(a) with the ones feeding MeOH directly to the column (Fig. 4) shows that conversion levels are similar. For low conversion

levels at low pump around ratios, we observe a slightly better performance for configurations with side-reactor feeds (e.g. approximately +2% for  $R_{\max} = 1$ ). For high pump around ratios, the difference in conversion is vanishingly small (e.g. approximately +0.3% for  $R_{\max} = 10$ ). This indicates that feeding pure MeOH directly to a side reactor is only beneficial as long as the chemical driving force in the reactor gets raised. For large recycle flow this is not the case. Distributing the MeOH feed might also be favourable with regard to hardware design, since a feed to the column could be replaced.

Furthermore, we tried to increase conversion by moving the feed from the column to the side reactors. Since the HOAc feed is also used for extractive distillation in the rectifying section, we were not able to detect a relevant increase in conversion when feeding part of HOAc directly to a side reactor. Furthermore, most of our attempts resulted in lower conversion.



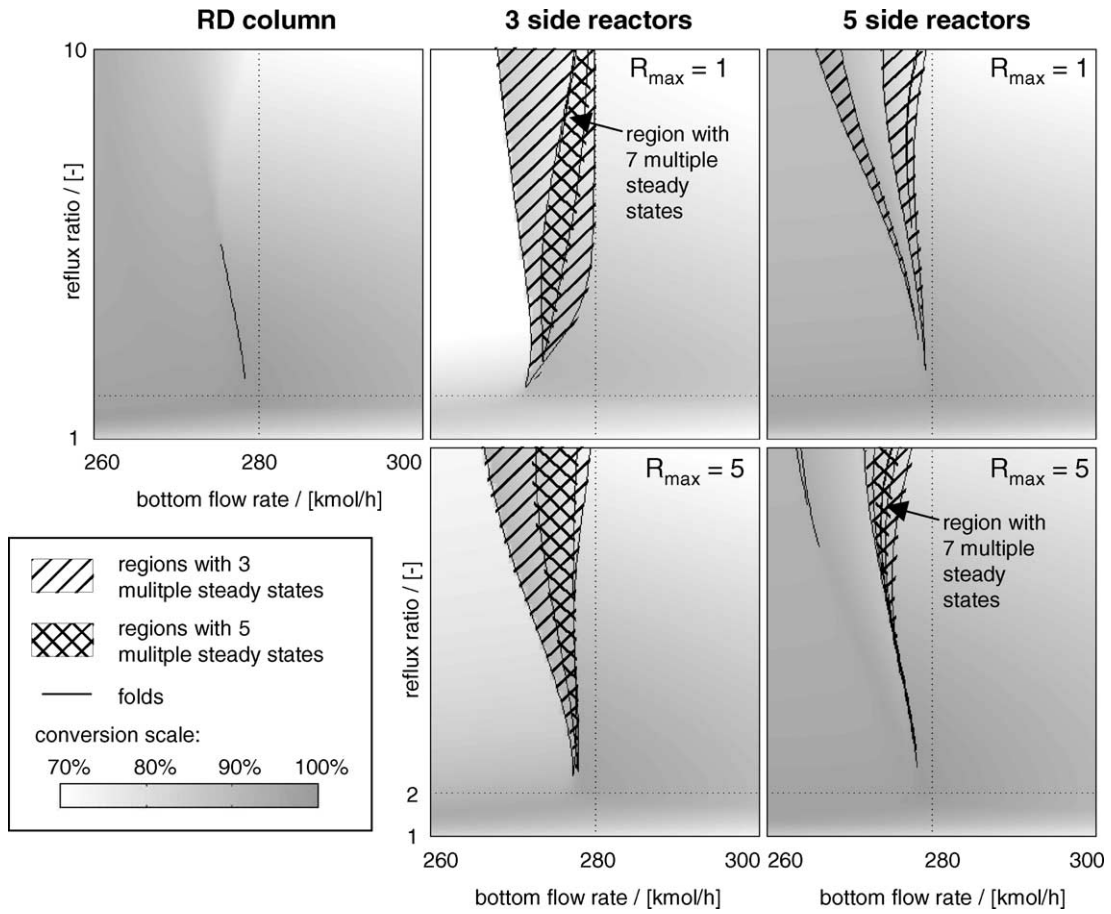


Fig. 8. Regions of multiple steady states for the RD column and for configurations with three and five side reactors.

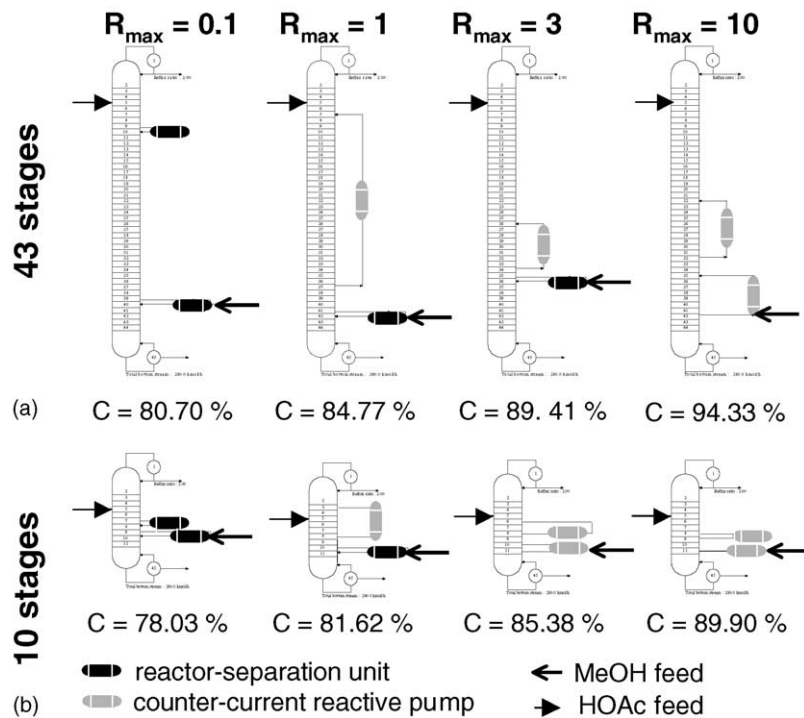


Fig. 9. Column configurations with two side reactors for (a) 43 and (b) 10 stages. MeOH is fed to one of the side reactors.

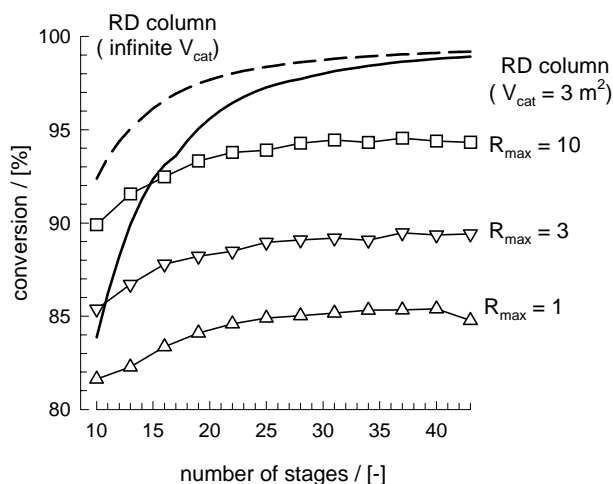


Fig. 10. Diagram HOAc conversion with respect to the number of stages. The reflux ratio is two and the bottom flow rate 280 kmol/h.

## 7. Number of stages in the distillation column

A major incentive for the side-reactor concept is to avoid in situ catalyst-loading limitations. In contrast to a column with side reactors, the catalyst load of an RD column is directly linked to the number of stages. In order to illustrate

this, we determined column configurations for various numbers of stages. For column configurations with two side reactors, we fed MeOH directly to one of the reactors. Fig. 9(a) and (b) shows typical configurations obtained for 10 and 43 stages. The RD column did not contain inert stages and its MeOH feed location was optimised in order to obtain high conversion. The HOAc feed was fixed at Stage 5 for all configurations.

Fig. 10 shows the attainable conversion for several column configurations when the number of stages is varied. As expected, reducing the number of stages yields a drop in conversion for RD and side-reactor configurations. In case of RD, this decline is much more pronounced for configurations with finite catalyst mass than for configurations with infinite catalyst mass. The reason for this is that reducing the number of stages not only results in poorer separation, but also in a narrower reaction zone. Hence, the overall catalyst load is implicitly reduced too. This effect does not apply to the side-reactor concept. The overall catalyst mass located in the side reactors remains constant albeit the number of stages in the column. Therefore, Fig. 10 reveals for side-reactor configurations that conversion equivalently declines. Hence, the drop in conversion can be attributed to lower separation ability when stages are removed.

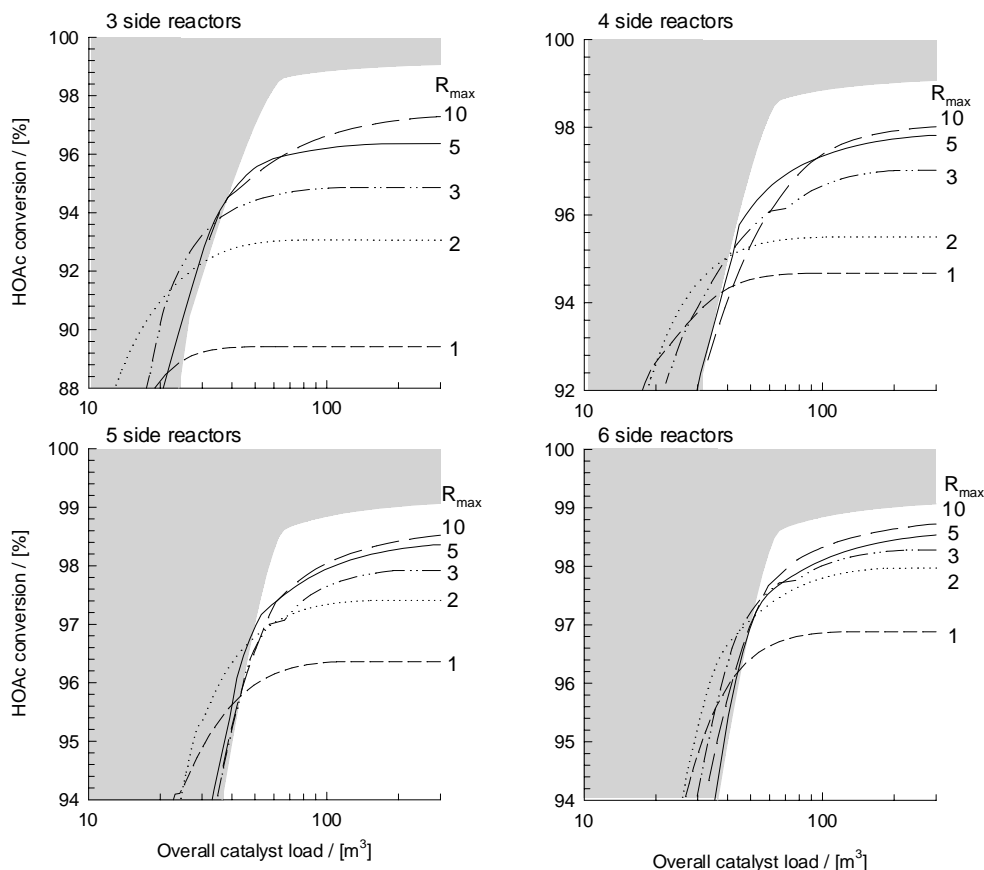


Fig. 11. Diagram of HOAc conversion with respect to the overall catalyst demand. The grey shaded area denote cases for which a side-reactor configuration would result in higher conversion with equal amount of catalyst. The configuration labels correspond to the ones in Fig. 4.

## 8. Catalyst demand

Fig. 11 shows the dependence of catalyst load on conversion. The grey shaded area denotes case for which a side-reactor configuration would yield higher conversion than a reactive distillation column with a catalyst load evenly distributed on each stage. The lines denote the side-reactor configurations as presented and denoted in Fig. 4. When previously designing the configurations, we assumed chemical equilibrium for the side reactors in order to determine the catalyst load. We modelled the side reactor as a plug flow reactor. For better comparison we also expressed the catalyst load of a side reactor in terms of liquid volume and used the same kinetics as in case of reactive distillation design. The initial catalyst load and distribution was estimated by minimising the catalyst load required in order to match at least 99.9% of reactor conversion at chemical equilibrium.

For the MeOAc process, if the target conversion is say 90%, even a 3-side-reactor configuration demands a lower catalyst load than an RD column. From the results presented in Fig. 11, we see that increasing the number of side reactors and the pump around ratio, we can match the catalyst demand of the RD column for any specified conversion level.

Fig. 11 shows the drop in conversion when the catalyst load in each reactor is proportionately reduced. For small catalyst loads, the side reactor concept appears to be superior to a reactive distillation column. This is caused by the advantage of using a plug flow side reactor. On the other hand the side-reactor concept is not attractive if high purity and conversion specification have to be met. Note, in the present case study, purity and conversion are linked since the two products are recovered at the top and bottom of the column, what makes it particularly attractive for reactive distillation. Furthermore, when comparing the catalyst demand of the side-reactor configurations with high and low pump around ratios, we notice that high recycle flows are not economical. As mentioned before, high recycle flow rates require high energy demand as well as low driving forces; consequently one requires significantly more catalyst load in order to reach close to chemical equilibrium (see Fig. 11).

## 9. Concluding remarks

In this paper, we have developed an algorithm to determine an optimum configuration of the side-reactor concept in order to maximise conversion. For the case study of MeOAc production, we see that it is possible to match the conversion level of an RD column by appropriate choice of the number of side reactors and the pump around ratio. The higher the conversion target the larger the number of side reactors and pump around ratios. For modest conversion levels, say

<90%, even a 3-side-reactor configuration will be able to match the performance of an RD column.

The study presented here reveals the potential, and limitations, of the side-reactor concept for use as an alternative to RD technology.

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## Appendix A. Nomenclature

$a_i$	activity of component $i$
$c_l$	total molar concentration in liquid (mol/m <sup>3</sup> )
$k_f$	forward reaction rate constant (s <sup>-1</sup> )
$K_{eq}$	equilibrium constant, dimensionless
$L$	liquid molar flow rate (mol/s)
$N_{RPA}$	number of reactive side reactors
$R_{max}$	limit for the pump around ratio
$T$	temperature (K)
$V_{cat}$	volumetric liquid holdup (m <sup>3</sup> )

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