

## Use of Compact Heat Exchanger as Flexible Reactor

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## Abstract

Compact heat exchangers (CHEs) are one of the most widely accepted Process Intensification (PI) devices in the chemical process industry (CPI). Highly compact heat exchangers with area densities in excess of  $10000 \text{ m}^2/\text{ m}^3$  can be manufactured by using micromachining and other modern techniques. A wide range of CHEs, from enhanced shell-and-tube to plate-and-frame and plate-fin heat exchangers to diffusion bonded heat exchangers are available. The classification of CHEs based on the hydraulic diameter has been somewhat arbitrary in the literature and no consensus has been arrived yet. For the purpose of this paper, heat exchangers with hydraulic diameter less than 6 mm have been classified as CHEs.

Use of such CHEs to carry out fast and highly exothermic chemical reactions can result into number of potential benefits such as substantially reduced inventory and environmental impact, higher levels of safety, improved temperature control, better product distribution and heat integration. Though, the use of CHEs to carry out such reactions was first envisaged about 25 years ago, CHEs specifically designed to carry out such chemical processes known as compact heat exchanger reactors (CHERs) have become available recently. These CHERs are capable of performing reaction, heat transfer and mixing (mass transfer) in a single unit. Considering the wide acceptance and well proven history of the CHEs combined with the potential benefits, the CHERs are likely to find application at industrial scale in near future.

Analysis of the potential benefits offered by the CHERs and the opportunities available in different sectors of CPI indicates that low tonnage sector - a large segment of CPI could be one of the biggest gainer by their application. However, it has been realised at an early stage that the application of CHERs for these processes is not straightforward. The majority of the chemical processes in low tonnage process industry involve complex multiple reactions with inherent uncertainties and different basic demands such as heat transfer, mixing (mass transfer) and hydrodynamics apart from large product portfolio and fluctuating demands for the products. Though state-of-the-art, present designs of CHERs are not likely to stand a chance against advantages offered by conventional reactors such as stirred tank reactor in dealing with inherent uncertainties and wide range of basic demands associated with such processes in real situations. Wider acceptance of CHERs in low tonnage process industry could be made possible by enhancing and exhibiting their abilities to closely match the basic demands for a range of reactions and to cope with the associated uncertainties.

One of the possible ways identified in this work to achieve this is to introduce more flexible ways to design, operate and configure CHERs. This paper proposes a novel approach to make CHERs more flexible and hence making them capable to meet the basic demands for a range of reactions. The paper also discusses different ways of achieving favourable mixing (mass transfer) and heat transfer rates along with preferred hydrodynamic flow regimes for the given chemical process by exploiting the ability of CHERs to add second reactant(s) on a passage by passage and stage-wise basis. Use of the capability of CHERs in order to achieve additional flexibility in terms of favourable temperature profiles by heating or cooling of reactant and product streams have also been

indicated. An experimental programme has been designed to prove the application of this approach.

## Introduction

Compact heat exchangers (CHEs) are characterised by having a high area density, which means a high ratio of heat transfer surface to heat exchanger volume. Benefits offered by CHEs in terms of efficient use of material, volume and energy in thermal systems have played crucial role in driving heat exchanger designs towards higher compactness and this trend towards ultra-compact designs are likely to continue (Mehendale, Jacobi, & Shah, 1999). Apart from these benefits, CHEs also offer multi-stream and multi-pass configurations, tighter temperature control, improved safety and radical approach to plant design. Highly compact heat exchangers with area densities in excess of  $10000 \text{ m}^2/\text{m}^3$  can be manufactured by using micromachining and other modern techniques. The classification of CHEs based on the hydraulic diameter has been somewhat arbitrary and no consensus has been arrived yet (Hesselgreaves, 2001; Mehendale, Jacobi, & Shah, 1999; Reay, 1994). For the purpose of this paper, however, heat exchangers with hydraulic diameters less than 6 mm have been classified as CHEs. A wide range of CHEs such as enhanced shell-and-tube, plate-and-frame, plate-fin heat exchangers and diffusion bonded heat exchangers are available in the market.

Assessment of the basic features and the advantages offered by the CHEs with regard to the concept and goal of Process Intensification (PI) (Jachuck, 2002; Stankiewicz & Moulijn, 2000) readily permits classification of CHEs as a PI device. Comparison of the acceptance of the CHEs in CPI with respect to other PI devices shows them to be one of the most widely accepted PI devices. However, limited choice for the material of construction, concerns about fouling and mechanical reliability, lack of standard design and operational guidance, lack of design codes and conservatism in the industry are often blamed for slower acceptance of the CHEs (Gibbard & Gough, 2001; Reay, 1994). Many of these adverse factors have been taken up as immediate research needs and considerable work has been done and substantially more is still in progress (Goldstein et al., 2003; Hesselgreaves, 2001; Mehendale, Jacobi, & Shah, 1999; Rumbold, 2003)

External coupling of a reactive system with a heat exchange system, such as buss loop reactor to control temperatures of both exo- and endo-thermic reactions has been practiced in the industry for a long time. Multifunctional reactors with internal coupling of reactive system and heat exchange system have also been used extensively in the process industry (Dautzenberg & Mukherjee, 2001). Stirred tank reactor and tubular reactor with heat transfer capabilities are examples of such multifunctional reactors. However, the use of CHEs to carry out fast and highly exo-/ endo-thermic reactions could result into much larger benefits compared to the conventional reactors due to much shorter characteristic length scales and higher surface area to volume ratio (Kirillov et al., 2001; Phillips, Lauschke, & Peerhossaini, 1997). Though, the possibility of carrying out a chemical reaction within the passages of a printed circuit heat exchanger was identified in the mid-1980s (Rumbold, 2003), CHEs specifically designed to carry out chemical reactions also known as compact heat exchanger reactors (CHERs) have become available recently. Some of the potential benefits often highlighted for the use of CHERs over conventional reactors are substantially reduced reactive inventory, significantly less environmental impact, increased levels of

safety, improved process control, better product distribution, less downstream processing and waste generation and possibility for heat recovery (ETSU, 2001; Hearn & McGrath, 1994; Johnson, 2000; Jones, 1996; Phillips, Lauschke, & Peerhossaini, 1997; Rumbold, 2003). Furthermore, the combination of low reacting inventory, effective mixing, short residence time and high heat transfer enable reactions to be performed with operating conditions considered unsafe for the conventional reactors (Green et al., 2001; Johnson, 2000). Considering the wide acceptance and well proven history of the CHEs combined with the potential benefits outlined above, CHERs are likely to find application at industrial scale in near future (Hugill, 2003; Jones, 1996; Rumbold, 2003).

## **CHE as a CHER**

In theory, any CHE with little modification can be used as a CHER. Either all the reactants can be introduced at one end and the unreacted reactants and products collected at the other end or some of the reactants can be added gradually, in controlled manner along the length of the heat exchanger. Heat of the reaction is added or removed by the suitable utility or other reactants, preferably as soon as it is required or generated. However, the effectiveness of a CHE as a CHER, relies heavily on the nature of the flow in the passages in which the reaction is taking place and its mixing (mass transfer) efficiency, in addition to its capability of removing/ supplying the heat of reaction. Any CHE, in order to qualify as a reactor, must be able to strike a judicious and flexible balance between the mixing (mass transfer) and heat transfer requirements of the chemical reaction. In other words, the rate of mixing (mass transfer) and the heat transfer must closely match the rate demanded by the chemistry of the reaction so as to allow the reaction to take place at its inherent rate. This imperatively means that the qualification as the best CHE (in terms of thermal effectiveness and heat transfer rates) does not necessarily qualify it as the best CHER.

Hearn and McGrath (1994) have already suggested that the heat-exchangers are dependent on macro-scale behaviour for effective heat transfer, however, progress of the reaction requires mixing at the micro-scale. Another study (ETSU, 2001) concluded that different CHE geometries have markedly different mixing efficiencies. A comparative study (Green et al., 2001; Johnson, 2000) of CHERs to carry out fast liquid-liquid reaction indicates that each type offers very different characteristics, not necessarily matching with the demands of the reaction. Another study performed to carry out highly exothermic reaction with static mixer reactor (Brechtelsbauer & Ricard, 2001) reveals their inability to cope with large heat transfer rates. It should be noted here that these conclusions are not necessarily directly applicable to all real process conditions. However, these differences in the characteristics of the different types of CHERs for the same reaction suggest that there exists an imbalance in the abilities of the CHER to meet the basic demands of a reaction and also that each type of CHER possess unique mixing (mass transfer) and heat transfer capabilities. It could be concluded here that better design of a CHER, requires a well thought out compromise between the opposite ends of the spectrum of mixing scales- heat transfer and mixing (mass transfer), in order to match the basic demands of the reaction.

## Application areas for CHER

A CHER can be classified as multifunctional reactor capable of carrying out chemical reaction, heat transfer and mixing (mass transfer) in a single piece of device. The most obvious potential application of CHER could be to carry out high temperature, intrinsically fast, exo-/ endo-thermic reactions, especially those involving relatively slow secondary series or parallel by-product forming reactions. These secondary by-product forming reactions are normally induced by temperature or concentration variations or in some cases by extended residence time in the reactor. The ability of CHER to remove (or supply) the heat of reaction at the rate equal to that of generation (or depletion) and mix the reactants at the time scale required by the intrinsic rate of the reaction allows the reaction to take place at its intrinsic rate and thus has the potential to result into vastly improved product distribution (Edge, Pearce, & Phillips, 1997; Johnson, 2000).

A broad class of important reactions carried out regularly in CPI have a high intrinsic reaction rate and relatively high heat of reaction. Examples of such reactions include nitration, sulphonation, halogenation, oxidation, polymerisation, neutralisation, alkylation, acylation, emulsification, amination, hydrogenation, dehydrogenation, reforming, ammoxidation, condensation and azo-coupling (ETSU, 2001; Hugill, 2003; Phillips, Lauschke, & Peerhossaini, 1997). Many such intrinsically fast and exo-/ endo-thermic reactions coupled with complex chemistry are often encountered in low tonnage process industry. This very fact makes CHER an important PI reactor for low tonnage processes. Low tonnage industry also represents a major segment of CPI and thus stands out as a major customer of CHERs. However, it must be admitted at this juncture that the application of CHERs in the low tonnage process industry is not straightforward. A delicate balance between the flexibility demanded by such industry and the flexibility offered by different types of CHERs needs to be struck.

Mixing (mass transfer), heat transfer and hydrodynamic flow regime requirements for low tonnage processes can vary over the full spectrum due to the wide range of chemistries used and multi-product nature of the industry. This adds an important additional criterion of flexibility (offered in terms of mixing (mass transfer), heat transfer and hydrodynamic flow regime) to be considered while designing and selecting the reactor for low tonnage process industry. The solution often proposed to overcome this flexibility criterion is to have a jacketed stirred tank reactor (STR) equipped with a variable speed drive mixing system and the facility to use different types of impellers. In accordance with this notion of flexibility, majority of the reactions in the low tonnage chemical industry are run in the batch or semi-batch mode in the STR (Hearn & McGrath, 1994; Hugill, 2003). However, the STR fails to provide the essential uniform molecular scale environment for fast, multiple and exo-/ endo-thermic reactions and also pose a major hazard due to the presence of large reactive inventories (Jones, 1996). A number of other factors, including the change in business models, stringent environmental regulations, change in the acceptable levels of safety and availability of options are forcing the low tonnage chemical industry to resort to more efficient reactors than STR.

Use of high efficiency continuous tubular flow reactors with good control of reaction temperature profiles and mixing (mass transfer) times for carrying out such reactions can prove to be useful in alleviating the limitations of the STR to a large extent. In certain cases,

it would be inappropriate to use conventional tubular reactors due to their comparatively large throughput and lack of precise control of the progress of the reaction compared to the requirement of the low tonnage process industry. However, recently introduced meso- and micro-scale CHERs hold great promises in overcoming the limitations of the STR and to suit majority of the requirements of the low tonnage process industry.

### **Low tonnage process industry and flexibility**

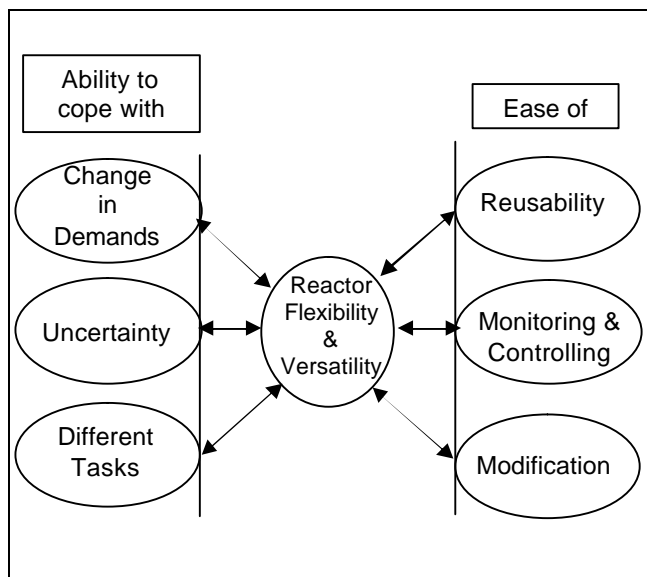
It is worthwhile to understand the needs for the flexible reactor for low tonnage process industry before exploring the flexibility offered by the CHERs. In this industry, one product is made, often in a campaign for a period that may last for days or few months. Then, the same plant with addition or deletion of a few of the processing components is used to manufacture another product. Reaction conditions and demands, reactants and also the inter-connection between the processing components may change drastically compared to that of the previous product. Also, to add to the complexity of the situation, new products with different processing requirements are continually added to the product portfolio due to short life cycle of the existing products. The overwhelming majority of low tonnage processes are of this nature, that is, a single reactor and upstream or downstream processing system components producing a portfolio of final products in a short time span. A major problem in the design and selection of the system components is that the substantial part of the demand that they are required to meet is often unknown at the start.

In such cases, a variety of process task sequences and poorly defined process requirements have to be considered. Similarly, the allocation of process tasks and sequencing of the components need to be changed frequently and quickly to match the new process requirements. In such situations, flexibility and versatility of the components including the reactor become important criteria for selection. The flexibility of any given reactor has been defined here as its ability to cope with the changes in the demands for different reactions, capability to handle the uncertainty inherent in the characterisation of the reaction and the capability to carry out other tasks than the reaction. The versatility of the reactor has been defined here as the ease of design modification to suit to the requirements of different reactions, the facility to monitor, control and adjust the reactions and also the reusability of the reactor for other purposes. The criteria summarized above can be used to judge the flexibility and versatility offered by different CHERs under consideration compared to that of the conventional STR (Figure 1). Criteria listed on the left in Figure 1 are more relevant to the term flexibility and the ones on the right to the term versatility. Also, the ability of the reactor to achieve quicker match with the demands of different types of reaction is likely to play major role in deciding its flexibility from the industry point of view. Apart from these flexibility and versatility criteria, the ability of exercising them quickly to match the demands of different reactions is also an important criterion for the selection of reactors in low tonnage process industry.

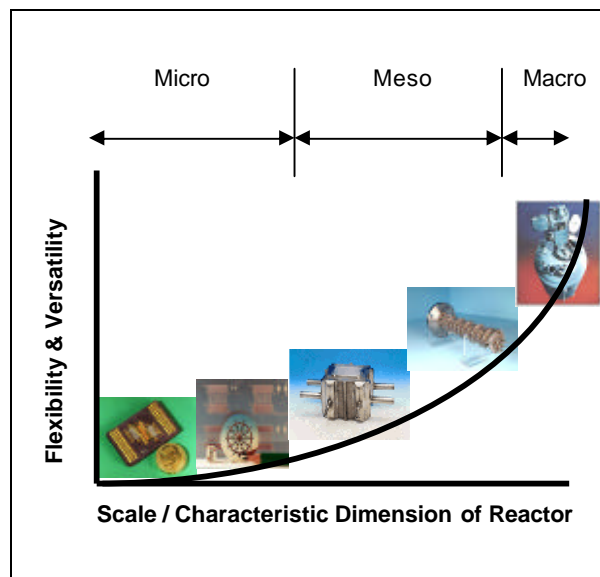
### **CHERs and Flexibility**

A wide range of CHERs such as shell and tube with static mixers (Flex-Reactor, Intensive Plug Flow Reactor), Diffusion bonded Marbond and Printed circuit reactor, Micro-reactor, Plate heat exchanger reactor and Plate fin heat exchanger reactor have been made available over the past few years. Suitability and even superiority of these reactors for

industrially relevant reaction schemes over conventional reactors have also been demonstrated at different levels (Edge, Pearce, & Phillips, 1997; ETSU, 2001; Green et al., 2001; Hugill, 2003; Johnson, 2000; Phillips, 1999). However, despite all the potential advantages associated with them, industrial applications of CHERs have been very limited so far. Normally, lack of experimental evidence, awareness, familiarity, standardisation, and reliability apart from industry conservatism are cited as barriers hindering the acceptance of CHERs at wider scale in the CPI (Hugill, 2003; Rumbold, 2003). However, one of the major reasons frequently omitted is the lack of proper characterisation of flexibility of the CHERs to cope with changes in process demands. Design, performance and application of the CHERs have been exhibited so far keeping in view very specific needs of the process and throughput, neglecting their flexibility aspects. This could have played a key role in spreading the perception about their inflexibility and making them unacceptable in the low tonnage process industry, where the flexibility holds prime importance. It is also a notion in the industry that the flexibility and versatility offered by the reactor decreases with scale of the reactor (Figure 2). CHERs would be accepted at much wider scale in low tonnage process industry if the perception about their inflexibility could be abolished and proved to match the flexibility and versatility criteria dictated by the inherent nature of the industry. It is often stated that the multi-stream, multi-pass capability of CHERs together with its ability to combine, mix and react different reactant streams in controlled manner in the optimum zone of mixing and heat transfer allows the mixing and heat transfer requirements of a range of reactions to be matched. However, experimental evidence and further clarifications to support such conclusions have not been presented.



**Figure 1 Flexibility and versatility criteria for reactor**



**Figure 2 Notion of flexibility and versatility of reactor as a function of its scale**

This paper introduces an approach to attain higher degree of flexibility with CHERs not only by design but also by configuring and operating them in novel ways. These additional dimensions for achieving the extended flexibility, favourably exploits the drastic reductions achieved in the size of the CHERs compared to conventional reactors. CHERs can be configured in a number of different ways which is not conceivable with conventional reactors. Due to substantially small reacting inventories and much more robust construction CHERs can also be operated in regimes considered unsafe with conventional reactors. These two novel and additional flexibility dimensions over and above the flexible design

bears great potential to make CHERs as flexible or probably more compared to conventional reactors. However, these three aspects of flexibility are not independent of each other and are likely to have a major influence on each other and also the overall flexibility. The importance of flexible design for CHERs can still not be underestimated and suggestions have been made to design them in more flexible ways.

### **Flexibility by Design**

CHERs with inherent flexibility are highly desirable as they could adjust themselves to cope with change in demands and tasks. They could also offer higher degree of tolerance to disturbances or uncertainties associated with the processing. The degrees of freedom available to design flexible CHERs can be divided mainly into two parts namely, Reaction side and Utility side. Likely degrees of freedom on the reaction side are number of feed/ exit points, feed point injection angle, adjustable distribution systems, flow path for secondary reactant, channel aspect ratio and surface properties. Likely degrees of freedom on the utility side are channel aspect ratio, flow patterns with respect to primary and secondary reactants such as co-, counter- or cross- and multiple entry/ exit points. It could also be possible to design CHERs that can be readily tailored to meet requirements of the given reaction with the use of adjustable features outlined above. Level of independency to adjust these features is also important as the effects are very much interwoven. Flexible design and ability to adjust the features of the given CHER independent of each other could also achieve decoupling between heat transfer, mass transfer and hydrodynamic regime to match the needs of certain reaction schemes.

### **Flexibility by Configuration**

It is possible to derive different types of useful configurations using similar or different types of CHERs to meet specific demands of reaction schemes over and above offered by the flexible design. Two of such plausible configurations have been illustrated in Figure 3. The configuration shown in Figure 3a could be used to carry out reactions requiring smaller mixing (mass transfer) time compared to that offered by the reactor itself. This can be achieved by configuring the reactors to offer simultaneous hydrodynamic and geometric focusing and thus reduced mixing (mass transfer) time (Shelat & Sharratt, 2004). This is often required for reactions involving viscous and non-Newtonian fluids where viscosity increases with the progress of reaction and the mixing (mass transfer) time has to be maintained same along the length of the reactor. The method of achieving focusing by configuration compared to the use of focused device provides greater degree of adjustability to suit the reaction requirements by using or bypassing the intermediate layers. By using the configuration shown in Figure 3b any variable residence time could be achieved while retaining the same flow pattern. Phoenix Chemicals, UK has successfully built a continuous equipment to carry out hydrolysis of ester product in aqueous environment using similar arrangement (Crystal Window, Issue No. 8, December 2003).

Other important aspects of flexibility offered by these arrangements are to control the introduction of the reactants, to adjust the mixing (mass transfer) time and to control the reaction temperature. Either all the reactants can be introduced at once at the first reactor layer or one of the reactant can be divided into number of portions and introduced separately at each layer to meet the concentration & temperature profile or hydrodynamics criteria depending on the reaction under consideration. Multi-step reactions involving use of



different reactants can also be carried out in one go using the availability of multiple feed positions.

### Flexibility by Operation

Elevated level of flexibility compared to that offered by the flexible design and configuration can be obtained by exploiting the different options of operation made available by the CHERs. Very small reactive inventories and ability to control the progress of the reaction precisely offers an extended range of process variables such as temperature, concentration and pressure compared to that offered by conventional reactors. Diverse conventional modes of operation such as semi-batch, continuous operation with or without recirculation can readily be obtained with CHERs configured and operated in appropriate manner.

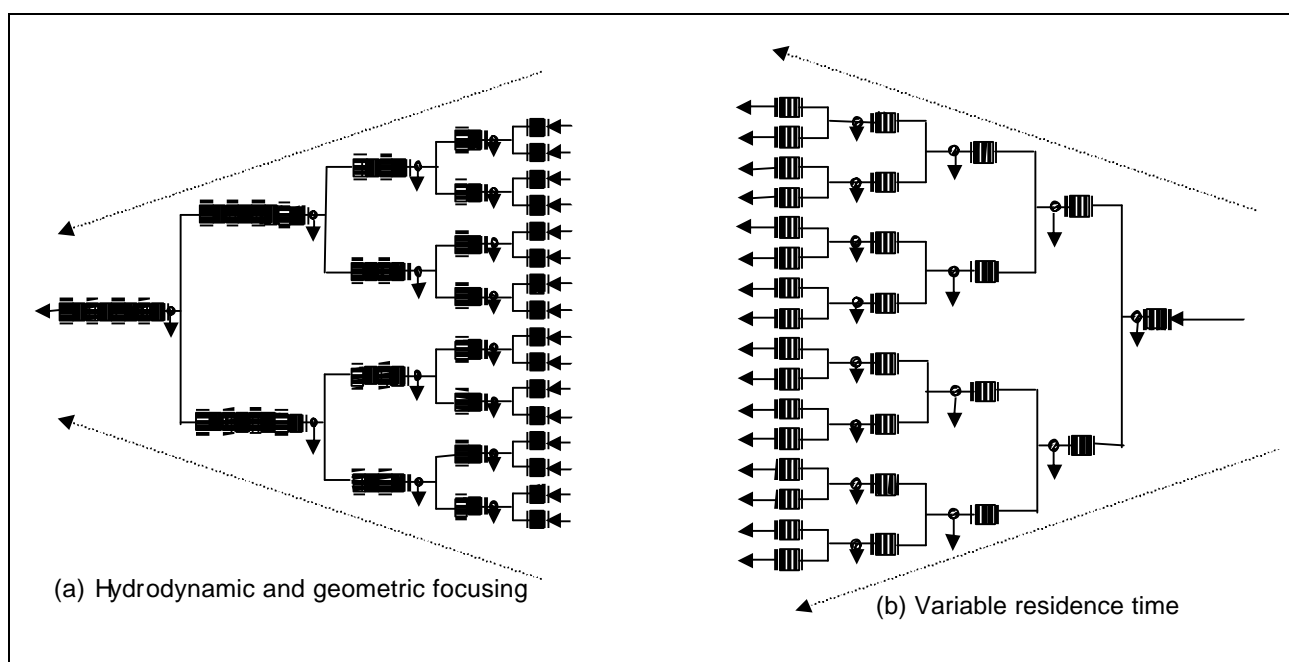
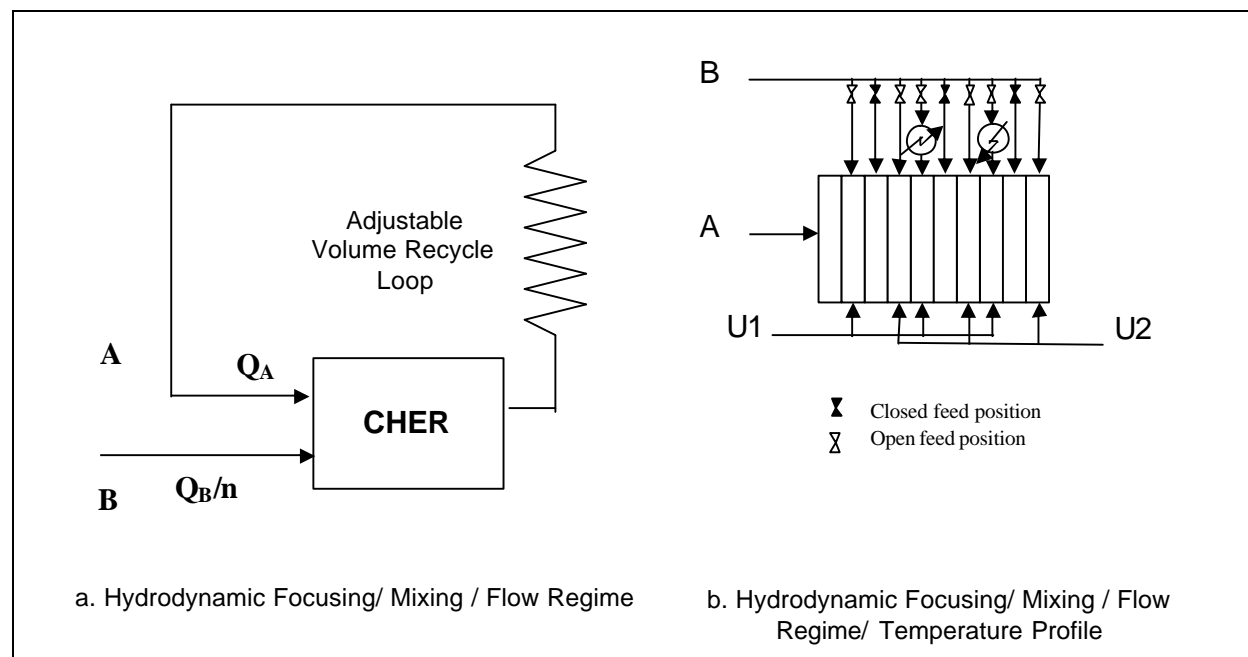


Figure 3 Flexibility by configuration

Figure 4a illustrates the use of adjustable volume recycle loop and addition of one of the reactants in multiple portions. The operation in this way could transform the reactor with single feed position into a reactor almost equivalent to one with multiple feed points. This method of operation can provide flexibility of hydrodynamic focusing, mixing (mass transfer), temperature control and hydrodynamic regime. As shown in Figure 4b the reactant B can be added at the predetermined positions and more than one utility can be used to control the concentration and temperature profiles along the length of the reactor.

As discussed above, it is possible to achieve improved flexibility and performance characteristics with the CHERs. However, close match between the heat transfer and mixing (mass transfer) characteristics of the CHER and the demands of the reaction is still the most important factor determining the product distribution for the fast, exo-/ endo-thermic and complex multiple reactions. In order to achieve the best possible product distribution for the given reaction, it is of prime importance to identify the possible design,

configuration and operating regime options offered by the CHERs beforehand. Without having this capability of identifying the options in the early stages of decision making process, the flexibility and improved mixing (mass transfer) and heat transfer characteristics offered by the CHER can not be utilised at the maximum and the results thus obtained could undermine the true potential of the CHER. To assist in the process of identifying the options at the early stages of decision making, it is vital to have a consistent and established model. A pragmatic model based on the analysis of the experimental results reported in the literature so far for the fast multiple reactions with both the turbulent and laminar flow reactors has been developed for this purpose (Shelat & Sharratt, 2004).



**Figure 4 Flexibility by Operation**

## Conclusion

The advantages and drawbacks of the CHEs that are also relevant to CHERs have been described. Benefits of using CHERs over conventional reactors such as STRs to carry out fast, exo-/ endo-thermic and complex multiple reactions have been highlighted. Features essential to use a CHE as a CHER have been stressed. Useful findings of the studies performed on the CHER to characterise their capabilities have been highlighted and features to be incorporated while designing the CHERs have been suggested. Potential application areas for CHERs with candidate processes have also been discussed. Factors responsible for the lack of enthusiasm about CHERs in particular and PI reactors in general at the industrial scale have also been outlined. The term flexibility in context of low tonnage process industry and the necessity of the flexible CHERs to match the demands of processes carried out in such industry has been explained.

Some of the key aspects of flexible design for CHERs in specific and PI reactors at large have been identified here. As has been shown, it is possible to configure and operate CHERs to derive additional flexibility over and above that offered by its design. However, designing flexible or adjustable CHERs is the most important step, as the flexibility offered

at this level is likely to determine the level of flexibility that can be achieved at other two levels. A judicious blend of these three levels of flexibility can enable CHERs to carry out a range of complex multiple chemical reactions having very different basic demands. Important questions including how to use flexible design, configuration and operation offered by CHERs to meet the demands of the given set of reactions still need to be addressed.

An experimental setup have been designed and constructed at the chemical engineering department, UMIST, UK to verify the proposed approach and exhibit the different levels of flexibility for the range of CHERs. Experimental proof of the approach is likely to pave a way in increasing the acceptance level of CHERs in low tonnage process industry in particular and CPI at large.

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\* Pictures have been used here only to represent the scale of device more clearly and have been taken from the websites <http://www.bhrgroup.co.uk/pi/flexreactor.htm>, <http://www.bhrgroup.co.uk/pi/hexreactor.htm>, <http://www.imm-mainz.de>.