Application of Printed Circuit Heat Exchanger Technology

within

Heterogeneous Catalytic Reactors

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Introduction

The Printed Circuit Heat Exchanger (PCHE) is an established compact heat exchanger technology, originally invented as a result of research performed at the University of Sydney in the early 1980’s. Heatric was formed in Australia, in 1985, to commercialise the concept (1), and the first applications were in industrial refrigeration systems. However, it soon became apparent that one of the biggest potential markets for the new technology was in offshore gas processing, where space and weight savings are at a premium.

Following Heatric’s move to the UK in 1989, PCHEs rapidly gained acceptance in the offshore industry, and some of the most significant early projects that used them have been extensively reported (2)(3)(4). Other applications include LNG, ethylene oxide, sulphuric acid, naphtha reforming, and caustic soda plants (5). A total of over 500 PCHEs have entered service around the world.

PCHE Construction

The compact core of a PCHE is constructed by chemically milling flow passages into flat metal plates, and then stacking and diffusion bonding the plates together into a single block. The chemical milling technique is analogous to that used for the manufacture of electronic printed circuit boards, and this gave rise to the “Printed Circuit” exchanger name.

Diffusion bonding is a high temperature solid state joining process that promotes grain growth across the metal boundaries, resulting in a join exhibiting parent metal strength and ductility.

If necessary, multiple diffusion bonded blocks may be welded together to form larger units, before headers and nozzles are welded on to complete the exchanger.

Alternatively, instead of welded-on headers, it is also possible to incorporate flow distribution channels within the diffusion bonded block, in a so-called “ported” design.

PCHE Characteristics

PCHEs can be up to 85% smaller and lighter than an equivalent shell-and-tube exchanger, as a result of the very high surface area density, high heat transfer coefficient, and potentially near-perfect counter current flow.
PCHE cores can be designed for extreme temperatures and pressures: units are in operation at over 500 bar (7000 psi) and temperatures ranging from cryogenic up to around 800°C (1472°F). They can be designed for very low pressure drops, even when handling highly viscous liquids or hot gas.

New Opportunities for PCHE Technology

The design and fabrication techniques developed for PCHEs can be applied in a range of new and novel areas. Examples include:

- mixer-reactors,
- cooling/heating with heterogeneous catalysis,
- cooling and flow distribution plates for fuel cells,
- cooling of electronics,
- reactions using supercritical solvents.

The potential for using a PCHE as a plug flow reactor was recognised by Heatric at an early stage. Since then, the mixer / heat exchanger / reactor concept has been widely investigated, and Heatric are currently working with partners to commercialise such devices: the program remains confidential, but we hope to be able to announce details at a future date. Fuel cell components are also an area of active development by Heatric. However, the primary focus of this paper is the temperature control of heterogeneous catalytic reactions.

Reactors for Heterogeneous Catalytic Processes

A wide variety of different reactor concepts have been used in heterogeneous catalytic processes, including fixed bed, moving bed, fluid bed, and bubble column. Within each broad category there are further variants. For example, a fixed bed may be arranged as one or more large packed beds, or may be packed within multiple tubes (with external cooling / heating, or with feed quench), or may be packed within the annulus of a double wall multi-tubular reactor. Each arrangement has been devised to address specific process requirements – especially temperature control. It is this challenge of
temperature control that is of particular interest to us.

The concept of using PCHEs in reactors is aimed at processes that are either highly exothermic or endothermic. Further, since PCHEs are suited to extremes of temperature and pressure, processes requiring such extremes are also potentially of interest.

Catalysts are commonly supplied on a porous support material such as alumina, inevitably resulting in poor thermal conductivity within the catalyst bed. Consequently, large fixed beds tend to operate adiabatically. Even in multi-tubular fixed bed reactors, commonly used for reactions involving large thermal loads - especially highly exothermic reactions, a very significant temperature gradient between the tube wall and the centre of the tube can arise. This poor in-tube heat transfer also frequently results in a highly non-isothermal axial temperature profile, with a pronounced hot spot close to the tube inlet. Loss of selectivity and product degradation are typical consequences of such uncontrollable hot spots, whilst thermal runaway and even explosion risk may occur in certain circumstances.

Such difficulties are sometimes addressed by adoption of some form of fluidised bed reactor but these can bring their own drawbacks, including high capital cost, catalyst loss (through attrition), possible flow instability or poor fluidisation, and risk of line plugging. They are also substantially back-mixed, which may be detrimental to yield selectivity in certain reaction systems.

Before looking at how we might use PCHEs to improve reactor designs, let's remind ourselves of the primary reactor design objectives. These are:

- to achieve an optimum temperature profile for selectivity and conversion;
- to minimize catalyst volume;
- to avoid catalyst overheating (with consequent irreversible damage);
- to minimize pressure drop;
- to achieve an optimal trade-off between capital cost and performance.

Application of extremely compact and highly flexible PCHE heat transfer technology allows us to address each of these objectives in new ways. We have adopted the term Printed Circuit Reactor or PCR to refer to such PCHE reactor applications.

“In Passage” PCR

The concept of coating a heat transfer surface with catalyst has been quite widely discussed: a recent study was presented at the 4th International Conference on Process Intensification for the Chemical Industry (8). However, one of the big challenges is how to provide sufficient catalyst surface at reasonable capital cost. The In Passage or IP PCR offers the desired high surface density at demonstrably competitive cost, over a range of operating conditions far exceeding that offered by any other type of compact heat exchanger.

The benefits of such a coated reactor are discussed in the case study given later in this paper.
Multiple Adiabatic Bed PCR

Inevitably it will not always be possible to match (at least approximately) the required catalyst surface with the required heat transfer surface. In this case the IP PCR risks simply becoming a rather expensive catalyst support, and an alternative approach is needed.

Where the required catalyst surface area is very large, and substantially exceeds the required heat transfer area, a better balance between capital cost and performance may be achieved by approximating the in-passage reactor with a large number of shallow adiabatic beds, with heat exchange between each bed – the Multiple Adiabatic Bed or MAB PCR.

A MAB PCR comprising an array of shallow catalyst beds interposed between thin PCHE panels is considered in the case study.

Alternatively, a very large number of small adiabatic beds can be incorporated within a single PCR block, together with heat exchange zones to adjust the initial feed temperature, to add or remove heat of reaction between each catalyst bed, and to adjust the product temperature. The heating or cooling medium may simply be a utility stream, or it too can undergo a separate reaction within a separate sequence of catalytic beds. Such an arrangement appears uniquely suited to reactors in applications where space and weight are at a premium – for example fuel reforming for automotive fuel cell systems.

Catalyst Considerations

With the “in-passage” reactor in mind, Heatric is developing and evaluating techniques for applying catalyst coatings to the passages within a PCR, and we believe robust and renewable coatings can be applied in a cost-effective manner. However, perhaps the biggest challenge is the availability of suitable catalysts. Substantially improved catalyst activity is needed if we are to take full advantage of the opportunity offered by a coated passage reactor. Catalyst life and resistance to poisoning or deterioration are also of paramount importance when considering layers only a few microns thick.
More encouragingly, as has been reported elsewhere \(^9\), both catalytic coatings and finely comminuted catalyst typically exhibit significantly improved effectiveness, when compared with commercially available pellets in a conventional packed bed. For a reactor with multiple small adiabatic beds contained within a single PCR block, a correspondingly small catalyst particle size is essential to assure good flow distribution and to avoid bypassing.

**Reactant Addition and Process Integration**

The well-established and proven technique for injecting glycol into offshore gas coolers can be extended to admix reactants into a process stream on a passage-by-passage basis. Further, it is possible to conveniently achieve staged reactant addition and mixing.

As previously noted, PCHEs offer the opportunity to combine multiple duties into a single heat exchanger, either sequentially or in parallel. Together with the capability for very high thermal effectiveness, these unique characteristics provide opportunities for significantly more compact and cost effective reactor designs.

**Case Study: Phthalic Anhydride**

**Background**

Phthalic anhydride (PA) is predominantly manufactured by the partial oxidation of ortho-xylene with air. The reaction is strongly exothermic (\(\Delta H = 1285 \text{ kJ/kmol}\)), and is carried out in a multi-tubular reactor cooled by circulating molten salt: the molten salt is in turn cooled by raising steam in an external boiler. The catalyst is \(\text{V}_2\text{O}_5\) on promoted silica gel, and the operating temperature range is 335°C to 415°C.

The following case study, based on a 50,000 tonne/year plant, shows clearly the significant potential capital cost and performance advantages achievable by using PCR technology.

**Conventional Reactor**

A typical multi-tubular reactor would comprise c. 28,000 tubes, of 3 to 5 m length, containing a total of 40 to 45 m³ catalyst. Indicative key operating parameters are:

- **o-xylene loading:** up to 100 g/Nm³ air
- **Conversion:** 70 to 71%
- **Selectivity:** 85%
- **Catalyst:** rings of 5 to 8 mm.

The capital cost of such a reactor is in the range of US$2.5-3.0 million.

**Multiple Adiabatic Bed PCR**

This concept achieves control of the reaction temperature throughout the reactor
by using multiple adiabatic beds alternated with heat exchangers in which the process stream is repeatedly cooled down to the optimum inlet temperature to the subsequent bed.

We have assumed the use of a structured ceramic matrix catalyst carrier, in order to minimize bed pressure drop, simplify catalyst loading and unloading, and maximize reactor compactness. In this case, gas flows along catalyst layers, instead of through the bed as in a traditional fixed-bed reactor: because the gas flows through straight channels the pressure drop is much lower than over a packed bed. Calculations show that pressure drop through structured catalyst is some 10 times less than through a fixed-bed. This saving can either be taken entirely as an energy credit (keeping the same superficial velocity as for a packed bed), or can alternatively be used to achieve a more compact, lower cost reactor (by increasing the velocity to give the same pressure drop as a packed bed).

Key benefits of such a configuration include:

- The PCHE panel face area matches the catalyst face area, so reactant redistribution problems, recirculating flows and dead spots are avoided.
- The PCHE and ceramic matrix structure both comprise passages with small dimensions – small enough to quench combustion or explosion.
- Any ortho-xylene feed loading is possible, from 44 g/Nm³ to more than 130 g/Nm³ in total. Indeed we believe it is quite feasible to operate entirely in a hydrocarbon-rich regime.
- Loading and unloading of catalyst is straightforward, as the whole matrix can be replaced in a very short time. In addition, as catalyst is prepared outside the reactor, there will be no need for adjustment of catalyst load (as frequently needed in each of the 28,000 tubes of a conventional reactor).
- The cooling medium flow pattern can be readily configured to ensure tight control over catalyst temperature profile, and if profitable, ramping the temperature upwards to compensate for reaction depletion in the later beds.
- PCHE elements are cost-effectively manufactured in stainless steel.

In this case study we have considered a PCR with 50 adiabatic beds and 50 PCHE elements, with ramped temperature profile. Each PCHE element is 4 m high and 1.1 m wide, to match a catalyst bed facial area of 4.4 m².

For 16,000 m² of structured catalyst surface, the catalyst volume will be 9 m³ and PCHE cores will have a total volume of 5 m³, so the total PCR volume, (excluding feed and coolant headers) will be 14 m³. This compares with an equivalent multi-tubular reactor volume of approximately 280 m³. This size comparison is illustrated in Figure 1. We have calculated a conversion of 72%, a selectivity of 78% for a total ortho-xylene loading of 120 g/Nm³.

The estimated cost of such a PCR, excluding catalyst, is approximately US$2.0 million. The overall operational
performance of this reactor gives potential savings of up to US$0.7-0.8 million per year.

Figure 1: Comparison between Multiple Adiabatic Bed PCR and conventional multi-tubular reactor.

Catalyst-Coated IP PCR

A catalyst-coated heat exchanger is clearly an attractive option for partial oxidation reactions, provided the coating:

- can be applied consistently throughout the structure;
- can be applied in a manner which sustains suitable levels of activity and adherence throughout a reasonable design life; and preferably,
- can be replaced or rejuvenated cost-effectively.

Catalyst-coated heat exchange surface can closely control reaction temperature, to an extent not possible with catalyst-packed tubes, as there is no large thermal resistance equivalent to that between the gas, catalyst pellets and the tube wall. Consequently the hot spot design constraint associated with packed tubes is largely eliminated and the average operating temperature of the reaction may be safely increased, subject only to constraints imposed by catalyst aging and undesirable side-reactions.

Key benefits of a coated IP PCR include:

- The reactant flow contacting the catalyst surface has no dead-spots or recirculating flows, which are known to compromise PA selectivity.
- The level of turbulence in the reactant flow can be readily adjusted to minimize gas-phase mass transfer resistances, which would become increasingly important at higher reaction rates. (We understand, on the other hand, that PA catalyst is relatively non-porous and pore diffusion is therefore unlikely to be important.)
- Process pressure drop can be readily optimized, trading off compressor power against reactor cost.
- By skillful application of counter-, cross- and co-flow, the cooling medium flow pattern can be configured to ensure tight control over catalyst temperature profiles.
- PCRs are cost-effectively manufactured in stainless steel.
- The PCR structure is inherently small-scale, and can be designed to quench any tendency for explosion. Therefore, there appears to be no fundamental constraint on ortho-xylene loading.
- The pre-heaters and post-coolers may be conveniently configured as PCHEs and close-coupled to the reactor, thereby minimizing piping and structure costs and avoiding the handling of explosive mixtures in large pipes.

Higher catalyst operating temperatures could well permit substantial reductions in required catalyst active surface. This effect could potentially be quite pronounced, since it appears the PA reaction rate roughly doubles for every 20°C temperature rise. For example, if the peak temperature at the center of a packed reactor tube were 50°C higher than the average temperature throughout the reactor, then conceivably only 25% of the
catalyst area would be required for a given yield of product in a catalyst-coated reactor.

For the capacity under consideration, the heat release in the reactor is approximately 16MW. Hence, if we consider the same 16,000 m$^2$ of catalyst surface as for the multiple adiabatic bed case considered above, the heat flux would only average 1kW/m$^2$. Clearly there is scope for at least an order of magnitude increase in heat flux, without overwhelming the heat transfer capabilities of the PCR core. Suitable ramping of the temperature profile could hold the heat fluxes relatively constant throughout.

Suitable ramping of the temperature profile could hold the heat fluxes relatively constant throughout. By making use of the full capabilities of the coated PCHE reactor, the capital cost may be brought down to around US$1.5-2.0 million, offering a significant saving over conventional technology.

**Other Options**

Reaction selectivity is adversely affected by the over-oxidation of PA to CO/CO$_2$. Yield improvements might therefore be achieved through options such as:

- a cascade of short-bed, low-conversion reactors with intermediate recovery of PA. PCHEs and PCRs are well suited to providing the required cascade of heat exchange surface cost-effectively.
- operation in the hydrocarbon-rich regime, perhaps with staged injection of air. PCRs are able to incorporate suitable means for intermediate reactant injection with intimate mixing. Enhanced safety would also be a benefit of operation under this regime if the mixtures were above the upper flammability limit.

**Conclusions**

PCHEs can be applied in novel reactor designs in several different ways, to achieve (near) isothermal conditions. Options include catalyst coating, or multiple adiabatic beds. Further possible features include staged reactant addition, and sophisticated heat integration.

Both the example tabled herein, and other ongoing design and test activities, indicate the use of PCHE technology can offer substantial economic benefits wherever:

- improved temperature control will give improved yields, or
- in instances where compact heat transfer can reduce capital cost, or
- wherever space and weight are at a premium.

In specific circumstances, further safety and utility cost benefits may also be realized.

**References**

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