CHAPTER 9 AREAS FOR FURTHER RESEARCH

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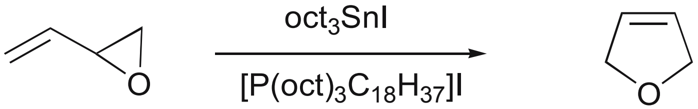
9.1 Introduction

A wide variety of new approaches to the problem of product separation in homogeneous catalysis has been discussed in the preceding chapters. Few of the new approaches has so far been commercialised, with the exceptions of a the use of aqueous biphasic systems for propene hydroformylation (Chapter 5) and the use of a phosphonium based ionic liquid for the Lewis acid catalysed isomerisation of butadiene monoxide to dihydrofuran (see Equation 9.1). This process has been operated by Eastman for the last 8 years without any loss or replenishment of ionic liquid [1]. It has the advantage that the product is sufficiently volatile to be distilled from the reactor at the reaction temperature so the process can be run continuously with built in product catalyst separation. Production of lower volatility products by such a process would be more problematic. A side reaction leads to the conversion of butadiene oxide to high molecular weight oligomers. The ionic liquid has been designed to facilitate their separation from the catalyst (see Section 9.7)



Equation 9.1 Commercialised Lewis acid catalysed production of dihydrofuran in a phosphonium ionic liquid.

In this chapter, we examine the various processes by taking a qualitative look at which parts need to be improved by further research in order to make them commercially attractive for the separation of lower volatility products and especially competitive with low pressure distillation. Once again we focus on the rhodium/tertiary phosphine catalysed hydroformylation of long chain alkenes, specifically 1-octene, since data concerning this reaction is provided in the preceding chapters. A summary of the best results obtained from each of the processes and the problems associated with their implementation appears in Table 9.1. A full economic analysis of each approach to the product separation problem is beyond the scope of this book, so any conclusions as to

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what research is still needed to render any of the processes commercially viable will at least to some extent be subjective.

9.2 Conventional separation methods (See Chapter 2)

Conventional distillation directly from the reaction chamber is not possible for longer chain aldehydes than C_5 because the volatility of the aldehyde is too low for it to be removed at an economically viable rate without raising the temperature above the decomposition temperature of the catalyst. In principle, an alternative to higher temperature is a faster flow of the gas phase through the reactor so that more product is removed per unit time. However, there is a limit in that liquid expansion (foaming) occurs and the reactor would have to be prohibitively large to prevent foam filling it entirely and the liquid catalyst solution also being carried out of the reactor. Once again, this limit is reached at about C_5 aldehydes. The formation of heavy (aldehyde dimer and trimer) products is also enhanced during the distillation process (higher aldehyde concentration and high temperature, see below).

Batch continuous processing, in which part of the catalytic solution is removed to a low pressure distillation unit, on the other hand, has recently been commercialised [2-4]. Very little information is available in the public domain concerning this low pressure distillation process, but the main extra cost will be in generating the reduced pressure required for the distillation. The estimated vapour pressures at 110°C of various long chain linear aldehyde products that are commercially desirable are shown in Figure 9.1. This temperature has been chosen because this is the high temperature limit above which the rhodium triphenylphosphine complex starts to decompose. Any commercial process will require to operate the product distillation step at a pressure no higher than those shown for the individual aldehydes.

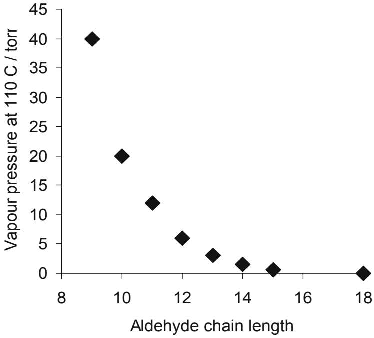

Figure 9.1. Estimated vapour pressures at 110°C of aldehydes of different chain lengths

TABLE 9.1. Rhodium catalysed hydroformylation of 1-octene using a variety of separation methods and catalyst systems compared with results from homogeneous commercial systems (First 4 entries). Modified from [5]

System	**Reference**	**p / bar**	**T / °C**	**TOF / h^{-1}**	**Rate / mol dm^{-3} h^{-1}**	**l:b**	**Linear aldehyde / %**	**Rh loss mg (mol product)$^{-1}$**	**P loss[a] mg (mol product)$^{-1}$**	**Problem[a]**
Homo / Co	[6]	300	150	38	3.2	2.3	50			
Homo / Co	[7]	80	200	20	1.6	9	80			
Homo[b]	[6]	15	95	770	2.0	8.8	90			
Homo[b]	[6]	16	110	444	0.8	5.3	83			
Supported	[8]	50	80	287	0.12	40	9.5	< 0.1	nr	
Supported / $scCO_2$	[9]	170	90	160		33	94	< 1.2	nr	R, P
Soluble polymer[c]	[10]	30	80	160	0.03	nr	nr	0.3	nr	R, S?, L
Dendrimer	[11]	10	120	1792	7.1	13.9	86	nr	nr	U
Supported Dendrimer[d]	[12]	67	45	25	0.03	0.05	92[e]	20	nr	R,L
Thermo-regulated Aqueous biphasic	[13]	50	100	182	0.3	nr	nr	nr	nr	R, S?
Fluorous biphasic	[14]	10	100	837	0.1	4.5	75	0.12	nr	S, L, E
Fluorous biphasic	[15]	20	70	4400	8.8	6.3	81	0.08	16	E
$ScCO_2$	[16]	200	65	430	14.2	5.5	79	< 0.17	nr	P
$ScCO_2$	[17]	125	100	162	2.5	3.2	75	0.014-0.07	nr	R, L, S
Ionic liquid	[18]	60	100	6200	5.3	44	85	< 0.003	< 0.06	E, I
Supported ionic liquid[c]	[19]	100	100	3600		2.4	71	0.004	nr	S, E?
$ScCO_2$ / Ionic liquid	[20] ([21])	200	100	517 (272)	7.8 (4.1)	3.1 (40)	76 (92)	0.002 (0.03)	nr	P, S, E (P, E)

[a] nr = not recorded, R = low rate, P = high pressure, S = low selectivity to linear aldehyde, L = high catalyst leaching, U = ultrafiltration not attempted, E = expensive ligand, and/or solvent, I high levels of isomerisation; [b] Rh/PPh_3 for propene, which cannot isomerise; [c]1-hexene; [d] vinyl acetate; [e] branched aldehyde; [f] leaching measured for styrene

The low pressure distillation may also lead to volatilisation of the ligand so that ligand losses may be high. Some of this ligand will be recycled, whilst some must be replaced. Finally, there is a major issue concerning heavies formation in systems manufacturing long chain aldehydes.

High boiling point products (heavies) are dimers or trimers produced from the aldehyde products by aldol condensation reactions. They, thus, have a molecular mass which is approximately twice that of the product aldehyde (or greater) and very much reduced volatility. In the production of butanal, they are removed at about the rate at which they are formed, so once they have built up sufficiently to act as the reaction solvent, they reach a steady state level in the reactor. For longer chain aldehydes, the heavy products are essentially involatile, so they must be removed by a liquid purge, which also removes catalyst. Here the separation problem is extremely severe. One way to reduce this problem is to operate in an inert diluent (reaction solvent). Since aldol condensation is second order in aldehyde (i.e the rate of aldol condensation α $[\text{aldehyde}]^2$), working at lower aldehyde concentration substantially reduces the rate of aldol condensation and the build up of heavies, thus allowing a longer time between liquid purges and a longer lifetime for the catalyst. Processes have been developed for the recovery of some of the catalyst in the liquid purge stream, e. g. by adsorption onto an insoluble support such as an acidic ion exchange resin [4].

Despite these problems, this low pressure distillation process has proven sufficiently economical to be commercialised for the hydroformylation of long chain alkenes and represents the benchmark against which all other processes must be judged.

9.3 Catalysts on insoluble supports (Chapter 3)

Until recently, all attempts that have been made to carry out catalytic reactions using metal complexes anchored to insoluble supports such as solid oxides or polymers have been prone to a degree of leaching that is too large for commercialisation, unless the leached material is recaptured on a guard bed of the same carrier, which in time takes on sufficient catalyst loading that it can be recycled as the active catalyst bed. The recapturing is carried out at lower temperature and pressure than the reaction, so that the stability constant for binding of the catalyst to the resin is maximised. More recently, catalysts have been developed in which the leaching is undetectable and single samples of catalyst have been used in a variety of reactions for over one year [22]. This is an exciting development, which relies upon the use of bidentate binding to the metal, so that the metal is retained with high efficiency, and covalent linking of the ligand into the solid matrix *via* a sol-gel reaction, ensuring that the ligand itself cannot break free from this support. The ligand shown in Figure 9.2 has been used successfully for the hydroformyation of 1-octene with no leaching being detectable (<0.005 ppm Rh, <0.1 ppm P).

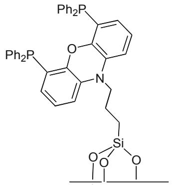

Figure 9.2. Ligand used for supporting a hydroformylation catalyst on a solid support. Leaching of rhodium under the reaction conditions is insignificant.

This is a very attractive process, which does not require any special engineering solutions because the catalyst can be used in a fixed bed, just like a heterogeneous catalyst. The cost of the ligand and the slightly diminished activity mean that cheaper more active catalysts of this kind represent a worthwhile goal and a significant challenge. Multipass operation may overcome the activity problem to some extent, but this has not been demonstrated.

Other factors which affect all new approaches to the catalyst separation problem include ligand degradation and heavies formation, although the latter may not be a serious problem for the supported catalysts as the heavy products should be removed with the organic product. In addition, there are the added complications of catalyst fouling and of mechanical abrasion that can occur with solid phase catalysts. The extent of these problems will only become apparent after continuous long term investigations under the conditions that would be used for a commercial process. The recent advances with supported catalysts suggest that the time is right to carry out these studies.

The same types of catalyst have been employed in 1-octene hydroformylation, but with the substrates and products being transported to and from the reaction zone dissolved in a supercritical fluid (carbon dioxide) [9]. The activity of the catalyst is increased compared with liquid phase operation, probably because of the better mass transport properties of $scCO_2$ than of the liquid. This type of approach may well reduce heavies formation because of the low concentration of aldehyde in the system, but the heavies that do form are likely to be insoluble in $scCO_2$, so may precipitate on and foul the catalyst. The main problem with this process, however, is likely to be the use of high pressure, which is common to all processes where supercritical fluids are used (see Section 9.8).

Supported aqueous phase (Chapter 3, Section 3.6, Chapter 5, Section 5.2.5) and supported ionic liquid phase catalysts, Chapter 7, Section 7.3) are probably not suitable for use with higher alkenes because the liquid feed slowly dissolves some of the water or ionic liquid changing the nature of the catalyst and leading to catalyst leaching.

9.4 Catalysts on soluble supports (Chapter 4)

Dendrimers and soluble polymers provide alternative supports to solids, which have the advantage that access to the catalytically active sites is not restricted. The main problem in these cases is not in the catalysis – reactions with high rates and selectivities have been reported - but rather in the separation which relies on nano- or ultrafiltration. One way of circumventing these problems is to support dendrimers on solid beads so that flow systems or simple filtration can be used. Because of the nature of the dendrimers, all their active sites are on the surface so all are accessible from the liquid phase even if they are supported on an insoluble support. This approach has only been applied in a very restricted number of reactions [12, 23] so increased research in this area could prove to be very productive.

For dendrimers and soluble polymers, there are two main problems: leaching of metal from the catalyst and lack of retention of the dendrimer or polymer by the membrane. Leaching could perhaps be addressed by using bidentate coordination and covalent attachment of the ligand, as in the solid supported catalysts described above, whilst the retention issue is potentially more difficult. The macromolecules could leak through the membrane either because the size of the channels is varied and some are large enough to allow the macromolecules through or because of imperfections such as pinholes.

Although membranes have successfully been developed for ultrafiltration in biological systems in water (removal of whole cells), the research on membrane materials for use with organic solvents and under gas pressure is in its infancy. This is an area where considerable further research could be carried out. Preventing membrane fouling is also an area where more information is required. Cross flow arrangements seem to be much more successful in this regard than dead-end filtration and they also allow continuous (loop) operation. Attention to the detailed shape and design of the channels could well pay large dividends in improving the efficiency of such systems.

Heavies that form in these reactions may be of sufficient size to be excluded by the membrane and hence retained with the catalyst. This would present a major problem as their separation from the catalyst will be non trivial.

9.5 Aqueous biphasic catalysis (Chapter 5)

Water - organic biphasic systems are the most developed of all the alternative technologies for homogeneous catalyst separation. Such a system has been commercialised by Ruhr Chemie/Rhône Poulenc/for the hydroformylation of propene to butanal and has been operating successfully for many years. The major problem with adapting such a process to the hydroformylation of longer chain alkenes does not concern the product separation, but rather the reaction itself. Very low rates are achieved using longer chain alkenes. Despite the fact that this approach was first described 30 years ago, it is still not entirely clear where the reaction takes place – in the aqueous phase or at the water – organic interface. It almost certainly does not occur in the organic phase or catalyst leaching would be a problem. Detailed studies aimed at clarifying this question would be highly beneficial.

As outlined in Chapter 5, Section 5.2.3.2 various approaches to overcoming the low rates of the hydroformylation of long chain alkenes in aqueous biphasic systems have been proposed. Some of these, such as the use of microemulsions [24-26] or pH dependent solubility [27], have provided improvements often at the expense of complicating the separation process. Perhaps the most promising new approaches involve the introduction of new reactor designs where improved mixing allows for much better rates of mass transport between the three phases (water, organic and gas) [28]. The use of thermoregulated systems [13] in which a temperature dependent phase transition makes the catalyst organic phase soluble under the reaction conditions, but water soluble on cooling is a very neat approach worthy of more development. Thermomorphic or latent biphasic polymers may also be important in this regard [29]. Further investment in these types of study would be very useful and could allow the commercialisation of such systems. The very attractive properties of water make this approach one of the most environmentally beneficial and many the problems associated with ligand degradation, long term catalyst stability and spent catalyst recycling have been solved (see Chapter 5, Section 5.3). Heavy aldehyde dimers should be removed with the organic phase and so will have to be separated from the product, probably by simple distillation.

9.6 Fluorous biphasic catalysis (Chapter 6)

It is generally considered that the major obstacle to the commercialisation of reactions employing the fluorous biphasic concept is the cost of the fluorous solvent and the cost of the ligand, which must contain very large amounts of fluorine to retain the catalyst within the fluorous phase. However, preliminary calculations (see Chapter 6, Section 6.14) suggest that careful handling of both the solvent and the ligand could reduce these costs to well below 1 € per tonne of product and that the costs here will be lower than those resulting from rhodium losses. However, this analysis does not take into account ligand degradation to give alkyl phosphines with all the problems, outlined in Chapter 2, Section 2.6 and Chapter 5, Section 5.3, that this brings associated with lower catalyst activity and selectivity as well as poorer catalyst retention. It should also be noted that the environmental persistence and very high global warming potential of fluorinated compounds may make their licensing for wide scale use problematic.

One advantage of the fluorous ligands, which also has implications for reducing the cost, is that the electron withdrawing fluorous ponytails increase the stability constants for complex formation and favour the linear product more than less electron withdrawing ligands. This means that high selectivity towards the linear product can be obtained at low ligand:metal ratio (low ligand loading). Because hydroformylation reactions are inhibited by excess phosphine ligands, the low ligand loadings possible in these systems mean that high rates are obtained and the reaction can be run at lower temperatures, still giving commercially attractive reaction rates [15]. The lower reaction temperature should lead to less degradation of the ligand by all the possible mechanisms, so that ligand losses will be greatly reduced.

Heavy products will be dissolved in the organic phase so that contamination of the catalyst phase and regular purging will not be necessary.

One major problem remains, however, leaching of the ligand and catalyst into the organic phase. The calculations in Chapter 6, Section 6.14, assume that the rhodium leaching can be reduced to 1 ppb and that the ligand losses may be 10 times this. At present, no catalyst has been reported that comes anywhere near this goal, so significant research is required to discover new catalyst designs which retain the ligand much more effectively into the fluorous phase. In addition, the leaching is much reduced if reactions are carried out to high conversion [30]. The reactor design proposed in Chapter 6, Section 6.14, for obtaining high conversion is rather simplistic and research into new reactor design, which would allow high conversions under continuous flow operation would be highly beneficial.

9.7 Reactions involving ionic liquids (Chapter 7)

As indicated in Section 9.1, one of the few examples of the implementation of an alternative strategy for homogeneous catalyst – product separation involves an ionic liquid. In this reaction, the isomerisation of butadiene oxide to dihydrofuran (Equation 9.1), the product is sufficiently volatile to be distilled form the reactor under the reaction conditions in a continuous manner. Where this is not the case, the reaction kinetics require that the substrate and the catalyst both dissolve within the ionic liquid. If the product is much less polar than the starting material, it may phase separate and be removed by decantation. If, however, as in long chain alkene hydroformylation, the product is more polar than the substrate, it is likely that it too will be soluble in the ionic liquid and hence might require extraction. In practice, the engineering solutions described in Chapter 7, Section 7.4 rely upon the ionic liquid becoming saturated with product so that a separate organic phase consisting of unreacted substrate and products forms and can be separated by decantation rather as in the aqueous biphasic process. The main advantage of the using an ionic liquid over using water is that the ionic liquid can be tuned to dissolve the substrate, even if it is of relatively low polarity so that high reaction rates can be achieved. The main disadvantage in a chemical sense is that the ionic liquid may be soluble to some extent in the organic phase, thus leading to the leaching of both ionic liquid and catalyst. The catalyst can be anchored within the ionic liquid phase by making it highly polar, although not so polar that it does not dissolve in the ionic liquid, but rather little has been reported about the solubility of the ionic liquids in the product stream. If the catalyst is retained within the main ionic liquid phase, the extracted ionic liquid can, in principle, be collected from the bottom of the product distillation column. In reality, this could be a major problem as the aldol condensation products, which are also essentially non-distillable, will be almost impossible to separate from this extracted ionic liquid. The cost of the ionic liquid means that such losses must be kept to an absolute minimum, so ionic liquids which dissolve the organic substrate but are insoluble in the product would be the ideal solution to the problem, although whether such liquids represent a realistic possibility remains to be seen. Most of the heavies will be in the organic phase, but they may also build up in the ionic liquid, in which case a purge will be required. Fortunately, on this occasion, the heavies should be extractable from the ionic liquid and catalyst using an organic solvent and the ionic liquid phase could be recycled.

In the Eastman process for 2,5-dihydrofuran production, the situation is different and the problem of heavy products has been tackled in a highly original manner.[31] The oligomers formed in the process are highly polar and insoluble in alkanes. The ionic liquid, $[P(oct)_3C_{18}H_{37}]I$ and the Lewis acid catalyst, $[Sn(oct)_3]I$, which are non toxic ($LD_{50} > 2$ g kg^{-1} for each), non-flammable (flammability 1) and non-corrosive (340 stainless steel is used for the reactor), have been designed to be soluble in heptanes. Part of the ionic liquid containing the oligomers, which is being recycled from the distillation unit to the reactor, is purged to a counter current extractor where the catalyst and the ionic liquid are extracted into heptanes. They are then stripped of the heptanes and returned to te reactor whilst the oligomers, now freed from the catalyst and ionic liquid, are incinerated.

Another solution to the problem of ionic liquid loss to the organic phase is to extract the product from the ionic liquid using a supercritical fluid (See Chapter 8, Section 8.2.2.3). It has been demonstrated that this can be done continuously for a variety of reactions including the hydroformylation of long chain alkenes [20], and that neither the ionic liquid nor the catalyst are leached to significant extents. The only problem here is the high pressures involved (see section 9.8).

Other problems with ionic liquids in addition to their cost, which is falling, are:

- their environmental persistence, especially important if they are lost with the heavies purge
- their lack of flammability, which may mean that destruction by incineration is difficult
- their slight but significant toxicity [32, 33]
- and their corrosive properties [34]

Many of these problems may be overcome by using ionic liquids based on sugars [35] or deep eutectic mixtures. Deep eutectic mixtures such as that derived from choline chloride and urea (m. pt. 12°C [36]) or carboxylic acids [37] can be liquids and have very low vapour pressure. They have been successfully used as electrochemical solvents, but their use in catalysis remains little explored. Urea is a fertiliser and choline chloride (Vitamin B4) is a component of chicken feed so the mixture is environmentally acceptable.

With the myriad of ionic liquids that are available, there is surely ample scope for further research of these important solvents and considerable grounds for optimism that improvements of the current systems to an extent that encourages commercialisation are possible.

9.8 Reactions using supercritical fluids (Chapter 8)

Like water, carbon dioxide is and environmentally very attractive solvent. The ability to control its solvating power by simple swings in temperature and pressure makes it an ideal medium for homogeneous catalysis. The main problem compared with conventional systems is that such swings, especially in pressure, require costly recompression and care must be taken to control the temperature whilst the pressure swing is occurring so that mixed liquid and gas phases do not form. They are very difficult to pump. The results presented in Chapter 8 suggest that rates and selectivities for the hydroformylation of 1-octene and other long chain alkenes can be commercially attractive and that separation of the catalyst can be achieved at least in styrene hydroformylation using the CESS process [38] (see Chapter 8.2). These preliminary experiments on the CESS approach suggest that some rhodium leaching still occurs, so improvements will be required either in the ligand design or in the optimisation of the pressure swings to reduce this still further. The necessity to use costly fluorous derived ligands to render the catalysts soluble in $scCO_2$ adds extra impetus to the search for better catalyst retention. An attractive aspect of the CESS process is that the formation of heavy aldehyde dimers will be reduced because of the low aldehyde concentration and that any that do form will precipitate during the reaction and be easily separated. It is important, however, that they should not extract the catalyst from the $scCO_2$.

Biphasic systems in which the $scCO_2$ simply acts as a transport vector, but the catalyst is dissolved in a separate liquid phase mean not only that catalyst retention can be improved, but also that fewer pressure swings are required. The main disadvantage of these systems is that the solvent (water or polyethyleneglycol) can show some solubility in $scCO_2$, thus complicating the separation. Ionic liquids allow excellent, although not as yet perfect, catalyst retention and no loss of solvent [20]. Heavy products will be reduced because both the ionic liquid and the $scCO_2$ will act as diluents, but they will eventually build up in the ionic liquid phase. This will lead to a requirement to purge the ionic liquid – catalyst – heavies mixture, but once again, the heavies should be extractable with an organic solvent and the catalyst – ionic liquid recyclable. Their main disadvantage, which is shared by other liquid - $scCO_2$ biphasic systems is that the product must be extracted from the liquid phase and hence very high pressures (200 bar) are required. Once again, this increases the costs associated with the recycling of CO_2.

Much lower pressures (125 bar) can be used when the liquid phase, in which the catalyst is dissolved, is the steady state mixture of reactants and products that accumulates during the reaction, because the product simply has to dissolve not be extracted [17]. The lower pressure operation makes this approach particularly attractive, although heavies will build up in the catalyst rich phase and separation of the catalyst from the heavies will require processes similar to those developed for low pressure distillation (see Section 9.2).

Process designs for these types of system have been proposed [21] but once again careful control of temperature and pressure will be essential to avoid phase separation problems especially in the recycling gases (CO, H_2, CO_2). Two alternative approaches to simple decompression and recompression may be worthy of consideration. The first is simply to pass the decompressed gaseous phase over an oxidation catalyst so that any unreacted CO and hydrogen, together with any organic materials are oxidised to water and CO_2 and then to vent the CO_2. This would not increase the overall CO_2 burden very significantly because most of the CO_2 being discharged would simply have been discharged anyway if it had not been involved in the process. This completely gets around the recompression problem but the main losses would be of CO and hydrogen. An alternative to oxidation would be membrane separation, such as is used in reforming. In this way the CO and H_2 could be recycled separately from the CO_2 or the CO_2 could be discharged. Recompressing the CO and H_2 would be much less energy intensive than recompressing CO_2 because the amounts involved would be so much less and there would not be problems with phase separation.

The alternative approach draws form the experience of coffee decaffeination. The currently practiced process is carried out on a massive scale using $scCO_2$ to extract the caffeine from the coffee beans. The caffeine is not precipitated from the $scCO_2$ by decompression, but is extracted into water from which it can be separated. The great advantage of this process is that it can be operated at essentially constant pressure and very little energy is required for recompression of CO_2 in the recycling loop. It is difficult to see how such a process could be applied to truly homogeneous catalytic system, in which the catalyst is dissolved in the $scCO_2$, but it could be applied in systems where the CO_2 is the transport vector. In these cases, it would be necessary to pass the effluent stream from the reactor through a counter current of a solvent which dissolves the product, but is not itself soluble in $scCO_2$. An ionic liquid would be one possibility, especially in the cases where lower pressure operation ensures that the product partitions well into the ionic liquid (e.g. nonanal into 1-octyl-3-methylimidazilium bis(trifluoromethyl)sulphonamide at 125 bar [17]). The high solubility of CO_2 in ionic liquids of this kind will reduce the separation efficiency, but it may be possible to use other solvents which dissolve the product but not CO_2 and are not themselves soluble in $scCO_2$. The removal of the product from this solvent could then be carried out as described in Chapter 7.

9.9 Conclusions

None of the alternative strategies for catalyst/product separation has yet reached the point where it can be commercialised for the rhodium catalysed hydroformyation of long chain alkenes and there are very few examples of commercialisation in any catalytic applications. Batch continuous processing with low pressure product distillation has been commercialised but the complexity of the system suggests that alternatives may be able to compete.

In this book, we have examined the chemistry behind the possible new approaches to solving the separation problem and have discussed process designs that could be used to implement the Chemistry. In this chapter, we have addressed what we believe to be the biggest problems remaining to be solved for the different approaches described in the rest of the book and have attempted to provide pointers towards possible solutions. These should provide impetus for further research in this important area aimed at improving the chemistry, phase behaviour, differential solubility and process design and eventually lead to the commercialisation of many more homogeneous catalytic processes so as to make chemicals production much more environmentally acceptable.

9.10 References

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