CHEMICAL ENGINEERING RESEARCH IN THE NETHERLANDS

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INTRODUCTION

The process industry represents one of the most important economic activities in The Netherlands; more than half of the Gross National Product is to a large extent related to the chemical, petroleum, pharmaceutical, metallurgical and food industries. It is therefore not incidental that research in chemical engineering has a long and healthy tradition in The Netherlands. This tradition has been built up by several Dutch pioneer researchers. Kramers (Delft) is generally recognized as the father of physical transport phenomena, Van Krevelen (Staatsmijnen and Delft) was one of the first to lay out the theory of mass transfer with chemical reaction, now so firmly entrenched in the chemical reaction engineering literature; this theory has been successfully used by Dutch industry (Shell, DSM) for commercial exploitation of, e.g., acid gas treatment processes. Rietema (Eindhoven) provided seminal contributions to our understanding of fluidization and fluid mixing and has provided intellectual stimulus to several generations of scientists many of whom are now providing research leadership both within universities and industry. The contributions of Waterman (Delft) to process technology have been very influential. Zuiderweg (Shell, and later Delft) has provided several new insights into our understanding of distillation operation; this understanding has had a great impact within industry, not only within Shell in The Netherlands but worldwide. Van Deemter (Shell), Van de Vusse (Shell) and Van Heerden (Staatsmijnen), even from within industry have had a great impact on the thinking and language of chemical engineering as is witnessed by the citations of their early papers. Thijssen (Eindhoven) has made a lasting contribution towards the rigorous application of chemical engineering principles to food technology. Delft has a long tradition in industrial biotechnology (Kluyver, Beijerink and Kossen).

In the area of catalytic process research, Schuit (Eindhoven) has been highly influential; one talks nowadays of the Schuit school of catalysis. The early contributions of De Boer (Delft) in the area of adsorption are now classical. In the area of thermodynamics, the names of Van der Waals (Amsterdam) and Van Laar (Amsterdam) have left their indelible marks, while the name of Overbeek is inextricably linked with the development of the theory of surface phenomena. The Netherlands has a long standing tradition of excellence in the area of turbulence and fluid mechanics, an area in which Hinze (Delft) has made substantial contributions.

There are currently six departments of Chemical Engineering in The Netherlands located at Delft, Eindhoven, Twente, Groningen, Amsterdam and Wageningen. The research carried out at the universities has close links with industry. The closeness of the links can be traced *inter alia* to the fact that several of the professors in chemical engineering have had several years of industrial experience; this is in sharp contrast with the situation in the US, for example. There are several cases in which university research has had a direct impact on industrial operations (e.g. the products of university research work on amine treatment of acid gases have found direct industrial use in The Netherlands and abroad). Equally importantly, the PhD students trained at Dutch universities are able to use their academic expertise directly within the industrial environment. An emerging trend in The Netherlands is one of entrepreneurship; university trained PhDs see the direct relevance of their knowledge and are setting up consultancy organizations for marketing their know how. University professors continue to serve in various advisory capacities to industry.

The character of university research in The Netherlands, generally speaking, is carrying on in the firm tradition laid down by the pioneers. Increased internationalization and cross fertilization has implied that the research programme has broadened its base. Scarlett (Delft) brings with him the English school of particle technology and Marin (Eindhoven) has injected the 'kinetic thinking' of the Belgian (Froment) school. Krishna (Shell and, later, Amsterdam) along with Wesselingh (Delft and, later, Groningen) were instrumental in introducing a new approach to modelling of mass transfer processes; their Maxwell-Stefan school of thinking, which had its origins in UMIST, is already having its effect in education, research and industry.

A further trend which can be discerned is increasing attention to innovations. We shall cite just two examples here for illustrative purposes. Van 't Riet (Wageningen) has used hydrophobic and hydrophilic ultrafiltration membranes for fat separations in a very innovative manner. Van Swaaij's (Twente) work on a membrane reactor for the Claus process and the rotary conical 'quick contactor' are very likely to lead to commercially viable processes in the near future. Improved reactors and separation techniques and improved processes will be of vital importance in improving the competitive edge of the Dutch Industry.

One of the current governmental sources of research funding is the Netherlands Foundation of Scientific Research (NWO) and the Netherlands Foundation for Chemical Research (SON) Working Party on Chemical

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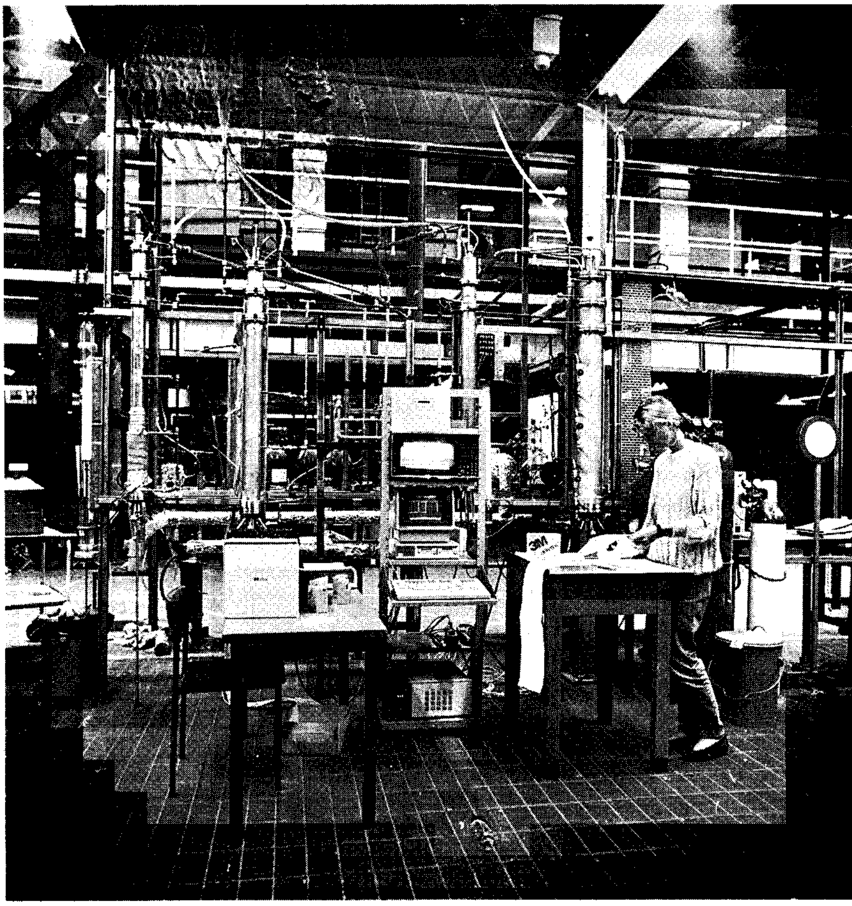

Figure 1. Pilot plant research facilities at the University of Twente.

Engineering plays an important role in the national coordination and stimulation of research. The STW (=Foundation for Technological Sciences) also plays a significant role in stimulating applied and innovative research. STW coordinates networks of scientists and industries and has several projects in chemistry and chemical engineering channelled via SON. A significant proportion of university research is directly funded by industry.

UNIVERSITY RESEARCH THEMES

The research activities at universities will be discussed under the following sub-headings: Thermodynamics, Transport Phenomena, Fluid Dynamics and Multiphase Flow, Colloidal and Interfacial Phenomena, Particle Technology, Chemical Reaction Engineering and Separations Engineering.

Thermodynamics

The group of De Swaan Arons (Delft) has several research themes including high pressure fluid phase equilibria and supercritical phase equilibria. Besides providing vital inputs to Dutch gas industry (Gasunie), the research efforts are directed at the development of novel separation techniques, e.g. supercritical extraction.

Transport Phenomena

Wesselingh (Groningen) and Krishna (Amsterdam) continue the development of the fundamentals of multicomponent mass transport phenomena using the concepts developed by Maxwell and Stefan more than a century ago. Their approach is being widely used within universities and industry, especially for membrane separations and separation processes design. The recently published text *Mass Transfer* (Ellis Horwood, 1990) has served to popularize the subject. Wesselingh's group concentrates currently on the determination of diffusivities in liquid mixtures and in macromolecular solutions and mass transfer under the influence of external fields (electrodialysis, ultrafiltration). Kerkhof (Eindhoven) is studying the transport processes in drying and in separations involving external fields. Moulijn (Delft) and Krishna (Amsterdam) are examining diffusion in zeolites, a subject which is important in several contexts within the process industries (adsorption, catalysis). Kerkhof (Eindhoven) and Van 't Riet (Wageningen) study transport phenomena in complex mixtures such as foods, sludge, clay and biomaterials. The use of micro-electrodes in the study of diffusion within biofilms is an interesting development (Ottengraf, Amsterdam; Luyben, Delft) and has applications in other areas. Hoogendoorn (Delft) has utilized the advanced theories of heat and mass transfer in multicomponent systems in the modelling of chemical vapour deposition processes. It is interesting to note that several of the classical assumptions made by chemical engineers (e.g. negligible thermal diffusion, negligible drift) are not valid for CVD processes. Van den Akker (Delft) will continue research on CVD reactors.

Fortuin (Amsterdam) is attempting to unravel the fundamental mechanisms of momentum transfer in turbulent flows. Such work is vital if chemical engineers wish to put their empirical correlations (e.g. Chilton-Colburn analogy) on a firm fundamental footing. Extension of such studies to include visco-elastic liquids will be highly enlightening. Janssen (Groningen) has recently focused attention on momentum transfer within boundary layers of visco-elastic fluids.

Westerterp (Twente) has focused considerable attention on almost all of the important transport process with gas-solid packed bed reactors; these studies have led to several new insights into the behaviour of packed beds, e.g. their 'statistical' character.

Fluid Dynamics and Multiphase Flow

The Netherlands has a long tradition of excellence in the area of fluid mechanics and this tradition is being maintained at several locations (Delft, Twente, Eindhoven). The recently formed J.M. Burgers Center for Fluid Mechanics at Delft has several themes of interest to chemical engineers, e.g. turbulent flows in process equipment, multiphase flows in fluidized beds and bubble columns, mixing and dispersion in stirred vessels, hydrodynamics of jets and sprays, cyclones, rheology and non-Newtonian flows, particle mechanics, modelling or turbulent flows with heat and mass transfer in combustors and furnaces.

In the area of multiphase flow, Van der Akker (Delft) is developing novel techniques for measuring locally and instantaneously bubble (swarm) velocities, bubble sizes, and bubble fractions both in gas-solid fluidized beds and in bubble columns. Computational multiphase fluid dynamics is receiving attention from Van Swaaij (Twente) and Van den Akker (Delft). Beek, Van den Akker and Van den Bleek (Delft) are investigating the non-linear flow dynamics of multiphase systems with a view to descriptions in terms of limit cycles and bifurcations.

In the area of two phase flow in pipelines, Fortuin (Amsterdam) has made several important contributions. Van den Akker (Delft) is investigating transient two-phase flows (pulsations) in pipe systems.

Surface, Colloidal and Interface Phenomena

The behaviour of colloidal particles in flow fields, which is of importance in precipitation processes and chemical reactions in emulsions (e.g. emulsion polymerization), has been the subject of experimental studies of colloid chemists (Stein, Eindhoven; Lycklema, Wageningen). Extensions of these studies to flow fields as encountered in stirred tanks have led to new insights with respect to scale-up (Thoenes, Eindhoven). Marangoni effects in distillation and extraction columns have been studied in the past by Zuiderweg and Stemerding. Janssen (Groningen) is pursuing a fundamental line of research on the influence of Marangoni convection on transfer coefficients.

An increased quantitative understanding of dispersions, gels, surfaces, micelles, electrical field effects will lead to new possibilities for the chemical engineer: separations with reversed micelles, new flotation techniques and electro-osmosis. There is considerable effort in several of the above areas amongst Dutch physical chemists (Frens, Delft; Stein, Eindhoven; Lycklema, Wageningen). Collaboration with these groups is growing; a welcome trend. Fruits of such collaborative efforts are evident in the work on reversed micellar extraction at Wageningen (Van 't Riet and Bijsterbosch).

Particle Technology

Scarlett (Delft) continues to focus on most of the important areas in particle technology. A fundamental understanding of particle technology is indispensable in our desire to produce structured materials and products (fibres, porous catalysts, membranes, etc.) using a variety of techniques (extrusion, spray drying, film blowing, etc.). Fundamental studies on particles will be valuable for the design of solids feeding devices, riser reactors, bunkers, etc., and absolutely essential in developing particle separation technologies. There is an increasing need in product technology for 'particle design'.

Chemical Reaction Engineering

This area is extremely well covered in The Netherlands and the coverage of topics is quite comprehensive.

Several research groups are involved in *kinetic measurements* and development of model 'reactors' (Marin, Eindhoven; Van den Bleek and Moulijn, Delft; Westerterp, Van Swaaij, Twente; Beenackers, Groningen). The determination of the kinetic constants for a complex reaction network is a research topic in itself and currently Marin (Eindhoven), Beenackers (Groningen) and Westerterp (Twente) have activity in this area. Heijnen (Delft) studies the kinetics and lumping strategies for complex biological systems.

Westerterp's sustained and continuing efforts in developing better understanding and design procedures for *packed bed reactors* have had considerable impact. Improved design procedures, improved correlations for heat transfer and dispersion and predictions of runaways have resulted from Westerterp's group at Twente.

There is considerable effort at several locations on *gas-solid fluidized beds* (Van Swaaij, Twente; Van den Bleek and Van den Akker, Delft; Thoenes, Eindhoven). Van den Bleek (Delft) is attempting to understand the different fluidization regimes and to improving scaling rules based on the chaotic behaviour of the fluid bed. Thoenes (Eindhoven) is improving available design procedures. Janssen (Groningen) is studying segregation and mixing effects in fluidized beds. Fluid bed combustion is receiving considerable attention at Delft (Van den Bleek, Moulijn) and Twente (Brouwers). Van Swaaij (Twente) has initiated studies on the hydrodynamics of dilute and dense phase (circulating beds) risers.

The idea of using fine adsorbent particles in *trickle flow through a packed solid bed* offers several interesting applications for catalysed reactions in which it is desirable to remove one of the products selectively (either to shift the equilibrium or to prevent side reactions). This concept has been demonstrated in practice at Twente (Westerterp and Van Swaaij). The remaining challenge is to scale up this reactor for commercial application (methanol?). The

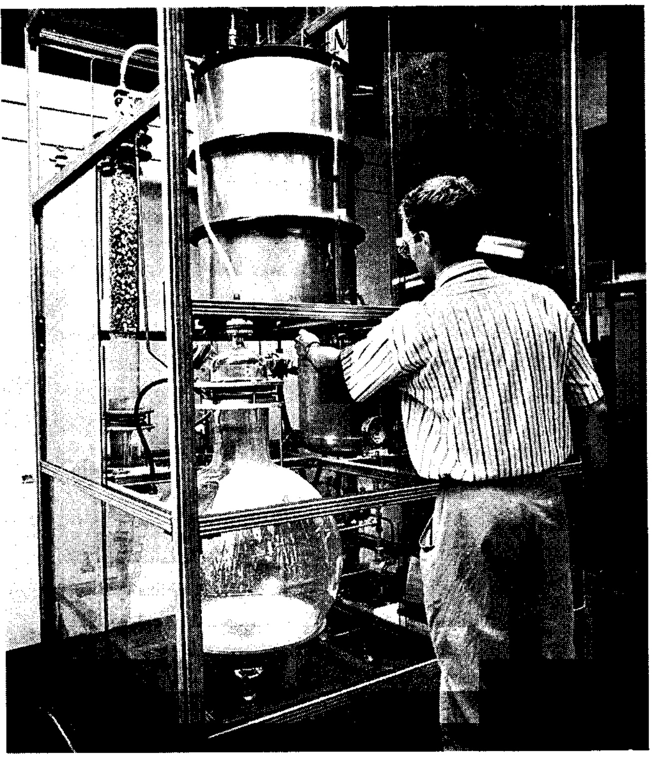

Figure 2. Gas treatment research at Twente.

solid trickle flow concept is being applied to acceptor processes for simultaneous treatment of SO_2 and NO_x (Van Swaaij, Twente, in collaboration with ECN).

Though there are literally hundreds of papers on *bubble column reactors*, a reliable correlation for gas hold-up for high pressure operation is still lacking. Industrial interest in this subject is great in view of the application of slurry technology for Fischer-Tropsch Synthesis of hydrocarbons. It is interesting to note that the first commercial unit for Fischer-Tropsch synthesis uses the multi-tubular trickle bed technology despite the fact that slurry technology is expected to be more cost effective. The reason for not choosing for the slurry technology in the first instance is the uncertainty with regard to the scaling up rules. A beginning has been made in the understanding of high pressure bubble column operation (Westerterp; Van Dierendonck) and this work needs to be extended further to slurry operations. It is interesting to note that the first fundamentally based model for gas hold-up under high pressure resulted from a collaborative effort between industry (Van Dierendonck, DSM), and university (Krishna, Amsterdam and Wilkinson, Groningen).

The *gas impelled loop venturi reactor* offers high mass transfer rates and is being studied by Beenackers (Groningen) and Van Dierendonck (DSM, Groningen). The analogous liquid impelled reactor system has several potential applications, e.g. in fermentations (Tramper, Wageningen).

For biotechnological applications, the *three phase air lift reactor* and a dual injection slurry reactor are being investigated at Delft (Heijnen, Luyben).

In the process of absorption of H_2S by iron chelates we have the interesting situation wherein fine sulphur particles are formed *in situ*, significantly enhancing mass transfer rates. This process is under study by Beenackers (Groningen). The influence of extremely fine particles on mass transfer is also being examined by Fortuin (Amsterdam) with a different process application in mind.

An area which requires increased attention from university researchers is the *three phase fluidized bed*, a promising candidate for the Fischer-Tropsch and Methanol processes. Beenackers (Groningen) is currently studying the fuel methanol process. In view of the complex interaction between the three phases, it appears necessary to undertake fundamental studies on sub-aspects of the problem. An interesting possibility is to operate the reactor with both coarse catalyst particles (i.e. three phase fluidized bed operation) along with fine adsorbent particles (slurry operation) wherein the adsorbent particles selectively adsorbs one of the reaction products. This four phase fluidized bed may offer advantages for some specific processes (Question: which ones?). Luyben (Delft) is studying the hydrodynamics of multistage fluidized beds with two suspended solids.

The appreciation of the analogies in the hydrodynamics of multiphase reactors may lead to new insights and better scale-up rules; this is the approach taken by Krishna (Amsterdam).

Though *trickle beds* are commonly used there are still several unknown factors with respect to design and scale-up, e.g. influence of high pressure on hydrodynamics and flow regime transitions; this subject is being studied by Westerterp (Twente).

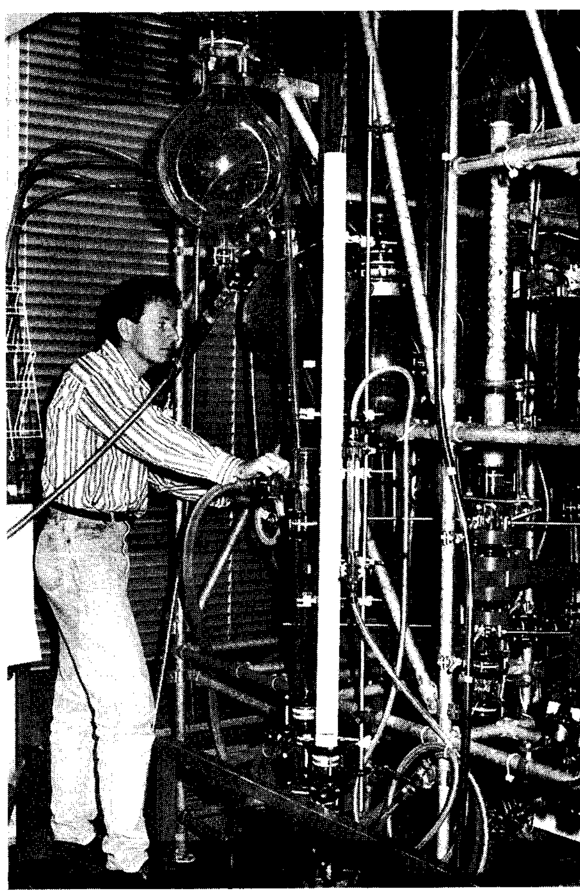

Figure 3. Studying the scale-up rules for pulsed pack columns.

Innovative Reactor Configurations

An alternative for the trickle bed reactor, the Bead String Reactor, is being developed at Delft (Van den Bleek). For gas phase reactions requiring strict stoichiometric control of the gaseous reactants membrane reactors offer interesting possibilities and advantages, e.g. the Membrane Claus reactor and membrane reactor for natural gas combustion (Van Swaaij, Twente).

Janssen and Beenackers (Groningen) are developing alternative reactor configurations (extruders, static mixers) for modification of starch; these offer possibilities of making new materials economically. Innovative reactor configurations will be required for tail pipe technology, i.e. off gas treatment processes requiring extremely low pressure drop. The ingenuity of the reactor engineer will be tested. Some progress has been made by Kema (in collaboration with Van Swaaij). The Bead String Reactor (Van den Bleek, Delft) is being developed as a low pressure drop reactor with a promise of improvement over commercially used parallel passage reactor of Shell. Unsteady operation of the reactors for NO_x conversion in stack gas could allow reduction of the required inlet temperature. Also, automobile exhaust gas conversion could profit from the unsteady operation of the converter (Moulijn, Amsterdam; Marin, Eindhoven).

New and innovative processes, e.g. double enzyme processes, are required to enable the production of optically pure chemicals (Heijnen, Delft).

There is increased interest in the use of homogeneous catalysts; a new research group in this subject has just been started at Amsterdam (Van Leeuwen).

Figure 4. Carrying out transient kinetic experiments at the University of Amsterdam.

In situ Separation and Reaction

There are several possibilities here:

- deliberate addition of an additional liquid phase to selectively extract the product and thereby prevent its further reaction, i.e. extractive reaction
- *in situ* distillation in the reactor, i.e. reactive distillation
- *in situ* adsorption; the reactor could therefore involve two solid phases (catalyst and adsorbent of different sizes. The question is whether one should keep one solid packed and the other phase in trickle phase flow (Westerterp; Van Swaaij), fluidize both phases (four phase fluidized bed)?
- *in situ* sorption and reaction; two components are removed from off gases: one by regenerative adsorption and the other by catalytic conversion. Van den Bleek and Moulijn (Delft) are studying such possibilities in a four-compartment fluid bed system and a rotation monolith.

Membrane reactors for selective removal of product can be thought of for:

- dehydrogenation reactions (dehydrogenation of ethyl benzene to styrene; propane dehydrogenation)
- fermentation (pervaporation membrane for removing butanol from fermentation broths (Luyben, Delft); continuous removal of micro-organisms, toxic products)
- hydrogen production via steam reforming (with selective hydrogen separation).

While the above discussions have been limited to *in situ* separation, the use of hydrides affords the possibility of *in situ* supply of reactant hydrogen (Van Swaaij, Twente).

Polymerization Reactors

Thoenes (Eindhoven) is examining emulsion polymerization in a continuous plug flow reactor. Janssen (Groningen) has a substantial ongoing activity in the use of extruders for carrying out polymerization reactions. For polycondensation reactions carried out within an extruder there are several challenges posed by the desire to remove (strip) the solvent *in situ*. There are several analogies here with reactive distillation.

Bioreactors

Ottengraf's (Amsterdam and Eindhoven) work on treatment of waste gases using biological filters offers interesting possibilities for pollution abatement. Luyben's work on the three-phase air-lift reactor may also be interesting for some petrochemical applications.

Reactor Stability and Dynamics

Westerterp (Twente) has an on-going programme of research on reactor problems related to hot spots, runaways and explosions. This research has provided vital inputs to industry. The availability of adequate experimental test facilities at Twente for determining the safe operating regions of chemical reactions is of immense use to industry. Fortuin's (Amsterdam) work on reactor dynamics and stability has attracted considerable interest from industry.

Expert system for Reactor Selection

The choice of the "ideal" reactor for given process requires careful attention in view of the substantial gains to be made in selectivity by use of novel configurations and *in situ* separations. In view of the vast number of reactor alternatives it becomes necessary to screen alternatives in a systematic way arrive at the selection of a reactor. Such a systematic methodology can be "coded" in the form of an expert system, which will be of immense value to industry and will also serve to point out gaps in our reactor research portfolio. Krishna (Amsterdam) and Van Swaaij (Twente) have embarked on a collaborative project, supported by industry, to develop such an expert system.

SEPARATIONS ENGINEERING

Distillation, Absorption and Extraction

As already mentioned above, The Netherlands has had a strong tradition of distillation research largely because of the efforts of Zuiderweg (Delft). These research efforts have had a considerable impact on industrial operations and some of the concepts and design procedures developed by Zuiderweg are now used worldwide. The unique, large scale distillation facilities at Delft are currently being used by De Graauw to study structured packed column hydrodynamics and supercritical extraction.

Fortuin (Amsterdam) is studying the hydrodynamics of the pulsed packed column for liquid-solid contacting; this apparatus has potential application in effluent treatment. Mass transfer in non-ideal liquid mixtures is being examined by Wesselingh (Groningen), who is using the Maxwell-Stefan theory in extractor design. Reversed micellar extraction is a new technique with potential applications in protein extractions. It has been developed in Wageningen by chemical engineers (Van 't Riet) working in close collaboration with physical chemists and biochemists.

Adsorption

Wesselingh (Groningen) is studying gel filtration and ion exchange chromatography. Kerkhof (Eindhoven) has an active group on adsorption from liquid phase with activated charcoal. Reith (Twente) is interested in adsorptive separations involving gaseous mixtures. The development of adsorptive separation processes using zeolites requires a better understanding of diffusion within zeolites, a subject under study by Moulijn (Delft) and Krishna (Amsterdam).

Crystallization and Precipitation

The subject of crystallization has been an important theme at Delft for more than two decades. The research group, initially led by De Jong, is currently under the leadership of van Rosmalen. A wide variety of research themes are receiving attention by the Delft group, which is one of the largest in the world. Precipitation and crystallization processes are also being studied fairly fundamentally by Thoenes (Eindhoven).

Drying

This area is important in, e.g. food, detergent and catalyst production industries; it receives continued attention at Eindhoven (Kerkhof). Van 't Riet (Wageningen) is examining transport phenomena, product quality and enzyme and micro-organism deactivation during the drying process.

Membrane Separations

The Dutch community owes a lot to Smolders (Twente) for developing membrane technology to its current high status. His group has been and is involved in just about every membrane separation process from gas separations to liquid membranes. Currently, development work on modules for membrane separations is underway at Twente (Reith). Kerkhof (Eindhoven) and Wesselingh (Groningen) are examining and developing a fundamental understanding of membrane separation processes. Van 't Riet (Wageningen) is examining fouling in ultrafiltration systems and the use of hydrophobic and hydrophilic ultrafiltration systems to obtain separations of emulsions. There is considerable experience in Wageningen on the use of membranes in bioreactors, all these efforts need to be supported and strengthened.

The area of ceramic membranes is also promising especially in view of high temperature processing and potential use in reactors; such membranes are being developed by Burggraaf (Twente), Geus (Utrecht), Van Bekkum and Moulijn (Delft).

Separations Involving Electric Fields

Ion exchange and electrodialysis are important for demineralization and desalting of aqueous and other streams. With continuing advance in membrane technology it can be expected that the applications of electrodialysis will increase. Wesselingh (Groningen) is looking at fundamental modelling aspects of electrodialysis. In Kerkhof's group (Eindhoven) subjects of study are scale rules for large scale electrophoresis, electrostatic coalescers and electric field driven gas dispersion and mass transfer in liquids.

Particle Separations

Separation of particles from gas and liquid streams is important and imposing increasing problems. The technology involved includes settles, cyclones, centrifuges, filters, flocculation, precipitation, electrostatic separation, filtration and dewatering equipment. Eindhoven (Thoenes and Kerkhof) has activity in the areas of filtration, precipitation and centrifugal flotation. Brouwers (Twente) has developed a novel rotational particle separator for separating liquid or solid particles of diameter 0.1 μm or larger from gases. Gas-particle cyclones are receiving attention at Delft (Van den Akker) and Janssen (Groningen).

There is a great need to increase our fundamental understanding of gas-particle and particle-particle interactions and in this connection the studies of Scarlett (Delft) are extremely vital.

RESEARCH SCHOOL IN CHEMICAL ENGINEERING

To increase further the utility of university research there is currently a move within The Netherlands to have a coordinated programme of research activities. It is planned to set up a Research School in Chemical Engineering which would be funded additionally by the Government and of which the existing Chemical Engineering Departments would be members. The basic idea is not to centralize all research activities but to further strengthen existing research groups in their own areas of excellence. There will be an effort made to avoid duplication of effort, especially in research areas requiring concentrated large amounts of inputs, e.g. pilot plant and large scale experimentation. The participants of the proposed Research School will be 20-odd professors of chemical engineering, along with some colleagues in related disciplines such as surface chemistry, combustion engineering and catalysis.

ADDRESS

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