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Four main objectives for the future of chemical and process engineering mainly concerned by the science and technologies of new materials production☆

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Abstract

Today the *chemical and process engineering* especially involving *chemical reactor engineering* has to answer to the changing needs of the chemical and related process industries such as petroleum, petrochemical, bituminous, pharmaceutical and health, agro and food, environment, iron and steel, building materials, paints, glass, surfactants, electronics, cosmetic and perfume, etc., and to meet market demands. So being a key to survival in globalisation of trade and competition, the evolution of chemical engineering is thus necessary. And to satisfy both, the market requirements for specific end-use properties of the products manufactured in (bio)chemical reactors and the social and the resource-saving and environmental constraints of the industrial-scale processes and technologies, it is shown that a necessary progress is coming via a multidisciplinary and time and length multiscale approach. In such a frame the future for the science and technologies of new materials can be summarized by four main objectives: (1) a total multiscale control of the process (or the procedure) to increase selectivity and productivity, i.e., nanotailoring of materials with controlled structure; (2) a design of novel equipment based on scientific principles and new operation modes and methods of production: process intensification; (3) product design and engineering: manufacturing end-use properties with a special emphasis on complex fluids and solids technology; (4) an implementation of the multiscale and multidisciplinary computational chemical engineering modelling and simulation to real-life situations: from the molecule to the overall complex production scale into the entire production site. Moreover, chemical and process engineering will also be increasingly involved and concerned with the application of *life cycle assessment* to new material design and production and its use but also to the plant and the equipment together with the associated services.

Keywords: Future of chemical engineering; New materials production; Multidisciplinary and multiscale approach; Triplet "processus–product–process engineering"; End-use property; Soft solids; Complex fluids; Data bank acquisition; Molecular modelling; Process intensification

1. Introduction: the moving world necessary requirements for chemical and related industries

The world moves forward. For the developing and industrializing countries, there is low cost of manpower and less constraining local production regulations. For the industrialised countries, there is a rapid development in consumer demand and constraints stemming from *public concern* over *questions of environment and safety*. In response to these changes, the world of chemistry and related industry including process industries such as petroleum, petrochemical, bituminous, pharmaceutical and health, agro and food, environment, textile, iron and steel, building materials, glass, surfactants, cosmetic and perfume, electronics, are confronted, from the technological and scientific point of view with a double challenge:

(a) *To research innovative processes for the production of commodity and intermediate products*. By no longer selecting processes only on the basis of economic exploitation but by seeking compensating gains resulting from the increased selectivity and savings linked to the process itself. This requires valorization of safety, health and environmental aspects, including the value of non-polluting

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technologies, reduction of raw material and energy losses and product and by-product recyclability as well. The industry will have to process with large plants supplying bulk products in large volumes. The customer will buy a process which is non-polluting, defect-free and perfectly safe.

(b) *To progress from the traditional intermediate chemistry to new specialities and active material chemistry and related industries*. This concerns industries involved with food products, with products for human, animal and vegetal health, with advanced materials, along with the chemistry/biology interface, i.e., the postgenomic world involving proteomics and metabolomics. This concerns also upgrading and conversion of petroleum feed stocks and intermediates, conversion of coal derived chemicals or synthesis gas into fuels, hydrocarbons or oxygenates. The aim is characterized by new market objectives, with sales and competitiveness dominated by the end-use properties of a product linked to its quality or shape and size (i.e., chemical and biological stability, degradability, chemical, biological and therapeutic activity, aptitude to dissolution, mechanical, rheological, electrical, thermal, optical, magnetic characteristics for solids and solid particles together with size, shape, colour, touch, cohesion, friability, rugosity, tasks, succulence, aesthetics, sensory properties, etc.). Control of the end-use property and expertise in the design of the process, its permanent adjustments to variety and changing demand along with speed in reacting to market conditions will be the dominant elements. Indeed for these new specialities and active materials the consumer buys the product which is the most efficient and the first on the market. He will have to pay high prices and expect a large benefit from these short life time and high-margin products.

Being a key to survival in global markets including the previous needs and challenges, chemical and process engineering necessitates the today evolution in teaching and revolution in research. Indeed the objective of petroleum engineering, then of chemical engineering broadened to process engineering, is the synthesis, design, scale-up or scale-down, operation, control and optimization of industrial processes that change the state, microstructure and (bio-agro)chemical composition of material through physico(bio)chemical separations (distillation, absorption, extraction, drying, filtration, agitation, precipitation, fluidization, emulsification, crystallization, agglomeration, etc.) as well as chemical, catalytic, biochemical, electrochemical, photochemical or agrochemical reactions. It involves the whole of scientific and technical knowledge necessary for physico-chemical and biological transformations of raw material and energy into the targeted products necessitated by the customer.

But it is important to note that today 60% of all products that a chemical company sells to its client are crystalline, polymeric or amorphous solids. These products need to have a clearly defined physical shape or texture in order to meet the designed and desired quality standards. This also applies to paste like and emulsified products. Instead of classical basic and industrial chemicals, new developments increasingly concern highly targeted and specialized materials, active compounds and special effect chemicals. These are much more complex in terms of molecular structure than classical chemicals and require a global approach for that manufacturing.

2. Le génie du triple “processus–produit–procédé”: the integrated multidisciplinary and multiscale approach of chemical and process engineering

Thus chemical and process engineering is now concerned with the understanding and development of systematic procedures for the design and optimal operation of chemical and process systems, ranging from microsystems to industrial-scale continuous and batch processes, as presented in Fig. 1 in using the concept of chemical supply chain [1]. This chain starts with chemical and other products that industry must size and characterize at the molecule level. Subsequent step aggregates the molecules into clusters, particles, and thin films as single or multiphase systems that finally take the form of macroscopic mixtures—solids, paste-like or emulsion products. Transition from chemistry or biology to engineering, one move to the design and analysis of the production units, which are integrated into a process that in turn becomes part of an industrial site with multiple processes. Finally this site is a part of the commercial enterprise driven by market considerations and demands involving product quality.

In this supply chain, it should be emphasized that product quality is determined at the micro- and nano-level and that a product with a desired property must be investigated for both structure and function. This involves a thorough understanding of the structure/property relationship at both molecular (e.g., surface physics and chemistry) and microscopic levels. And the ability to control microstructure formation to obtain the end-use properties of a fluid or solid product is the key to success and will help design and control product quality and make the leap from the nano-level to the process level.

This necessitates an integrated system approach for a multiscale and multidisciplinary modelling of the complex, simultaneous and often coupled momentum, heat and mass transfer phenomena and processes taking place at different time scales (10^{-15} to 10^{8} s) and length scales (10^{-8} to 10^{4} m) encountered in industrial practise (Fig. 2) involving approaches at the different length scales presented in Fig. 3.

So organizing scales and complexity levels in process engineering is now necessary to understand and to describe the relationships between events at nano- and microscales to better convert molecules into useful products at the process scale. And organizing levels of complexity by translating *molecular processes* (that I define by the name *processus*) into phenomenological macroscopic laws to create and to control

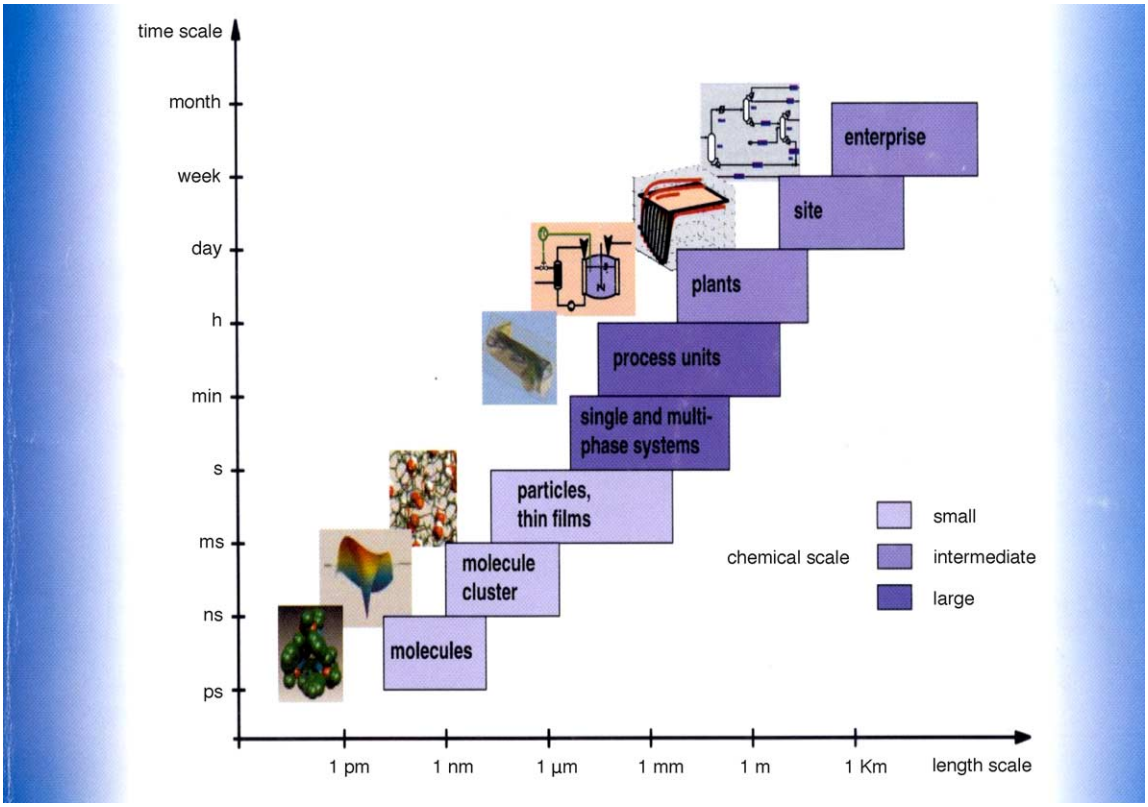


Fig. 1. Chemical supply chain [1].

the end-use properties and functionality of *products* manufactured by a continuous *process* underlie the today new views of chemical and process engineering. This can be defined by *le Genie du triple "processus–produits–procédé"* (the triplet molecular processes–product–process engineering, 3P engineering) with an integrated system approach of complex processes and phenomena occurring at different time and length scales [2].

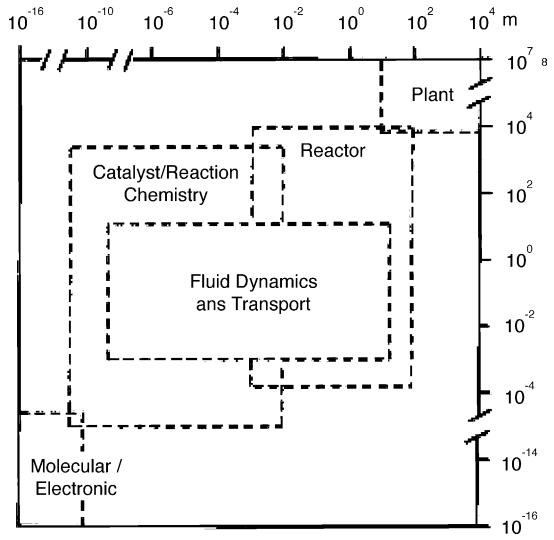


Fig. 2. The length and time scales covered in the multiscale approach.

This explains why, in addition to the basic notions of unit operations, coupled transfers and classical tools of chemical engineering, that is, in addition to the fundamentals of chemical engineering (separation engineering, chemical reaction engineering, catalysis, transport phenomena, optimization and process control), *this integrated multidisciplinary* and *multiscale approach* is a supplementary and considerable advantage for the development and the success of this engineering science in terms of concept and paradigms. This approach is of a great help in order to analyze, design and operate processes able to manufacture a product—first on the market—(often having a short life cycle) *with the desired property* and optimally thanks to *processes* involving if possible zero defect, zero pollution and zero accident.

And it will be possible to understand and to describe the relationship between events at nano-scale and micro-scale to better convert molecules into useful products at the zero pollution and zero accident process scale thanks to the large breakthroughs (a) in molecular modelling (both theory and computer simulation), (b) in scientific instrumentation and non-invasive measurement techniques (NMR, TAP, tomographic techniques, spectroscopic or monochromatic ellipsometry, i.e., diffusing wave spectroscopy, etc.) and related micro- and nanotechnologies in connection with image processing and (c) in powerful computational tools and capabilities, necessary for the treatment of generalized local information (increasing use of computational fluid dynamics

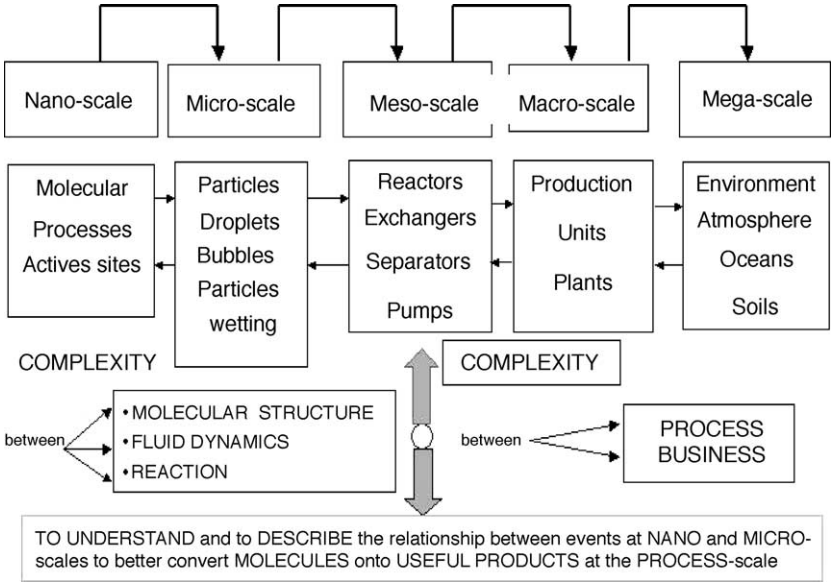


Fig. 3. Organizing levels of increasing complexity underlie new view of chemical engineering.

or mixing, e.g. CFDLIB, FLUENT, PHOENICS, FLOW 3D, FIDAP, FLOWMAP, etc.) [2].

3. Chemical and process engineering: quo vadis?

The previous general considerations on the future of *chemical engineering* concern four main objectives.

3.1. Total multiscale control of the process (or the procedure) to increase selectivity and productivity

This necessitates the "intensification" of operations and the use of precise nano- and micro-technology design. This is the case of molecular information engineering encountered for the supported organometallic catalysis or for supramolecular catalysis where instead of using porous support for heterogeneous catalyst, synthetic materials with targeted properties are now conceived and designed. Indeed, central to a successful catalytic process is the development of an effective catalyst which is a complex system in both composition and functionality. And the ability to better control its microstructure and chemistry allows for the systematic manipulation of the catalyst's activity, selectivity, and stability.

3.1.1. Nanotailoring of materials with controlled structure: opportunities for molecular engineering in catalysis

Indeed through the control of pore opening and crystallite size and/or a proper manipulation of stoichiometry and component dispersion there exists now ability to engineer via nanostructure synthesis novel structures at the molecular and supramolecular levels, leading to the creation of nanoporous and nanocrystalline materials [3] (Fig. 4). These materials both possess an ultrahigh surface-to-volume ratio, which offers a greatly increased number of active sites for carrying out catalytic reactions.

Nanocrystalline processing includes the tailoring of size-dependent electronic properties, homogeneous multicomponent systems, defect chemistry, and excellent phase dispersion. This provides nanocrystalline catalysts with greatly improved catalytic activity over conventional systems and multifunctionalities necessary for complex applications. For example for structure-sensitive reactions such as photocatalysis over titania used for decomposition of chemical wastes such as chloroform or trichloroethylene, catalytic activity depends not only on the number of active sites, but also on the crystal structure, interatomic spacing and crystallite size of the catalytic material. By varying crystal size and phase through molecular engineering, it is possible to manipulate and optimize the catalyst design of titania crystals of controlled size (4–100 nm) and phase which are systematically synthesized by sol–gel hydrolysis–precipitation, followed by hydrothermal treatment [3]. Specifically, 10 nm anatase crystallites due to their greater redox potential presents the best photonic efficiency for the photodecomposition of chloroform and trichloroethylene.

Also through supramolecular templating, nanoporous systems can be derived with well-defined pore size and structure, as well as compositional flexibility in the form of particles and thin films. Microporous materials including zeolites and tailored with well-defined pore structures for excellent surface areas and product selectivity are now typically derived through templating with individual molecules. The resulting zeolitic structure which consists of pore opening <1.5 nm allows only small molecules to enter and react, this providing shape and selectivity in separations and catalytic reactions.

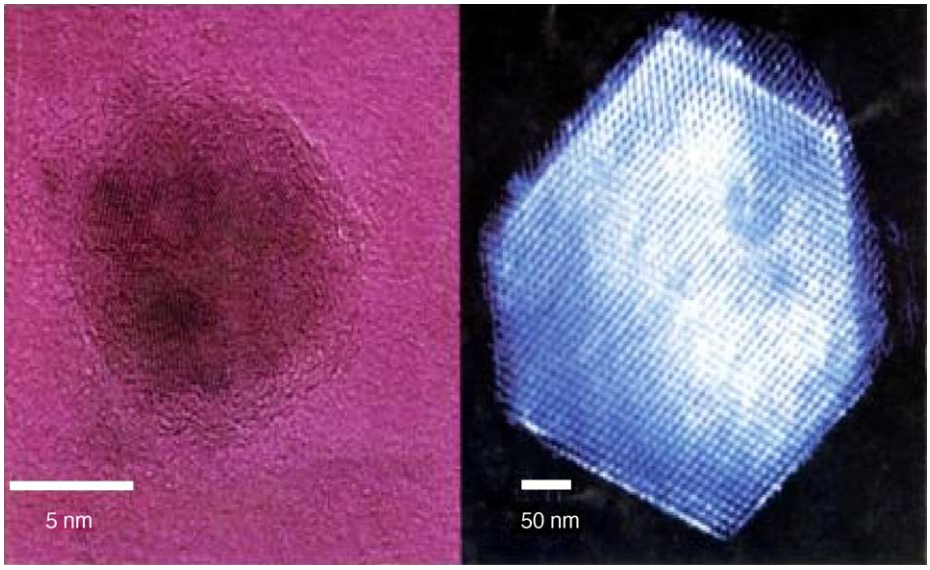


Fig. 4. Tailoring of materials with controlled structure [3].

Particularly noteworthy is the possibility of creating mesocellular foams produced by templating with triblock copolymers and trimethylbenzene. The resulting materials are composed of uniformly sized, large spherical cells up to 35 nm, which are interconnected by windows to create a continuous three-dimensional pore network. They are attractive for use as catalyst supports in pharmaceutical synthesis as they will permit the diffusion of large substrates through their large open-pore architecture. These porous matrices can also host oxide clusters and active metal and fixate organometallic ligands, offering new possibilities for creating heterogeneous catalyst useful in selective fine chemicals synthesis and asymmetric catalysis [3,4].

We could also add that in the field of homogeneous catalysis a supramolecular fine chemistry has been recently established extending the principle of self-organization of the enzyme (catalyst/molecule) to non-biological systems in using supramolecular compounds as catalysts for the shape selectivity of molecules. Such catalysts are formed in situ by self-organization, i.e., chemical bionics [5]. So the latest advances in nanotechnology have generated materials and devices with new physical characteristics and chemical/biochemical functionalities for a wide variety of applications. And chemical engineers and researchers are uniquely positioned to play a pivotal role in this technological revolution with their broad training in chemistry, physical chemistry, processing, systems engineering, and product design.

3.1.2. Increase selectivity and productivity by supplying the process with a local "informed" flux of energy or materials

At a higher microscale level, detailed local temperature and composition control through staged feed and heat supply or removal would result in higher selectivity and productivity than does the conventional approach, which imposes boundary conditions and let a system operate under spontaneous reaction and transfer processes. Finding some means to convey energy at the site (supplying the process with a local "informed" flux of energy) where it may be utilized in an intelligent way is therefore a challenge. Such a focused energy input may be achieved by using ultrasonic transducers, laser beams or electrochemical probes. And to drive the elementary processes within the unit is a challenge but combining microelectronics and elementary processes, e.g. tuning the selectivity by controlling catalytic reactions at the surface of electronic chips is a track being explored.

3.1.3. More clearly recognized is the necessity to increase information transfer in the reverse direction, from process to man

This means developing all kinds of intelligent sensors, visualization techniques, image analysis and on-line probes giving instantaneous and local information about the process state. This opens the way to a new "smart chemical and process engineering" requiring close computer control, relevant models, and arrays of local sensors and actuators. Field-programmable analog arrays coupled with microreactor technology promise to change the way plants are built, as well as the methods by which their processes are designed and controlled.

3.2. Process intensification: design of novel equipment based on scientific principles and new operating modes and methods of production

The progress of basic research in chemical engineering has led to a better understanding of elementary phenomena and now makes it possible to imagine new operating modes of

equipment or to design novel equipment based on scientific principles.

3.2.1. Process intensification using multifunctional reactors

Such is the case with the "multifunctional" equipment that couple or uncouple elementary processes (transfer–reaction–separation) to increase productivity, selectivity with the desired product or to facilitate the separation of undesired by-products. Indeed in recent years, extractive reaction processes involving single units that combine reaction and separation operations have received considerable attention as they offer major advantages over conventional processes: due to the interaction of reaction and mass and energy transfer, thermodynamic limitations, such as azeotrope, may be overcome and the yield of reactions increased. So the reduction in the number of equipment units leads to reduced investment costs and significant energy recovery or savings. Furthermore improved product selectivity leads to a reduction in raw material consumption and, hence, operating costs. So globally, process intensification through use of multifunctional reactors permits significant reductions in both investment and plant operating costs (10–20% reductions) by optimizing the process. In an era of emaciated profit margins, it allows chemical producers more leverage in competing in the global market place.

There exists a great number of reactive separation processes involving unit operation hybridisation.

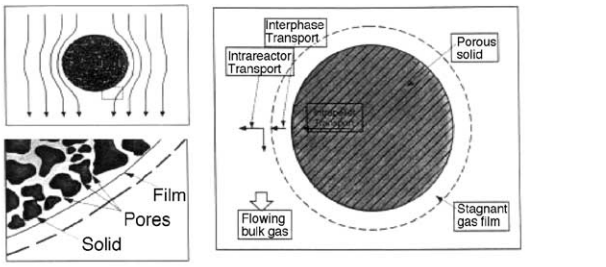
The concept of reactive or catalytic distillation has been successfully commercialized, both in petroleum processing, where packed bed catalytic distillation columns are used, and in manufacture of chemicals where reactive distillation is often employed. Catalytic distillation combines reaction and distillation in one vessel using structured catalysts as the enabling element. The combination results in a constant-pressure boiling system, ensuring precise temperature control in the catalyst zone. The heat of reaction directly vaporizes the reaction products for efficient energy utilization. By distilling the products from the reactants in the reactor, catalytic distillation breaks the reaction equilibrium barrier. It eliminates the need for additional fractionation and reaction stages, while increasing conversion and improving product quality. Both investment and operating costs are far lower than with conventional reaction followed by distillation. The use of reactive distillation in the production of fuel ethers such as *tert*-amyl-methylether (TAME) or methyl-*tert*-butyl ether (MTBE) or methyl acetate clearly demonstrates some of the benefits. Similar advantages have been realized with the production of high purity isobutene, for aromatics alkylation, for the reduction of benzene in gasoline and in reformate fractions, for the selective production of ethylene glycol which involves a great number of competitive reactions and for selective desulphurization of fluid catalytic cracker gasoline fractions as well as for various selective hydrogenations. The next generation of commercial processes using catalytic distillation technology will be in the manufacture of oxygenates and fuel additives [6].

An alternative reaction–separation unit is the chromatographic reactor, which utilizes differences in adsorptivity of the different components involved rather than differences in their volatility. It is, especially, interesting as an alternative to reactive distillation when the species involved exhibit small volatility differences or are either non-volatile and sensitive to temperature, as in the case, for example, in small fine chemical or pharmaceutical applications. In all these applications, special care has to be devoted towards the choice of the solid phase (sorption selectivity, sorption capacity and catalytic activity). Typical examples for the adsorbents used are activated carbon, zeolites, alumina, ion-exchange resins and immobilized enzymes [7].

Concerning the coupling of reaction and crystallization, there exist myriads of basic chemical, pharmaceuticals, agricultural products, ceramic powders, pigments produced by reactive crystallization based processes (i.e., processes that combine crystallization with extraction to solution mine salts). These separation processes are synthesized by bypassing the thermodynamics barriers imposed on the system by the chemical reactions and the solubilities of the components in the mixture. By combining crystallizers with other unit operations, the stream compositions can be driven to regions within composition space where selective crystallization can occur [8a].

The complementary nature of crystallization and distillation is also explored. Indeed the hybrids provide a route to bypass thermodynamic barriers in composition space that neither the distillation which is blocked by azeotropes and hindered by tangent-pinches in vapor–liquid composition space nor the selective crystallization which is prevented by eutectics and hampered by solid solutions and temperature-insensitive solubility surfaces, can overcome when used separately [8b]. Extractive and adductive crystallization are solvent-based techniques that require distillation columns. They are applied to high melting, close-boiling systems.

Membrane technologies respond efficiently to the requirement of so-called process intensification. Because they allow improvements in manufacturing and processing, substantially decreasing the equipment-size/production-capacity ratio, energy consumption, and/or waste production and resulting in cheaper, sustainable technical solutions. The paper by Drioli and Romano [9] documents the state of the art well involving progress and perspectives on integrated membrane operations for sustainable industrial growth. This technology can respond to the strongly increasing demand for food additives, feeds, flavors, fragrances, pharmaceuticals, agrochemicals, etc. Phase-transfer catalysis can also be realized in membrane reactor configurations, immobilizing the appropriate catalysts in the microporous structure of the hydrophobic membrane. Catalytic membrane reactors are also proposed to selective product removal to remove equilibrium limitations, i.e., catalytic permselective or non-permselective membrane reactors, packed bed (catalytic) permselective membrane reactors, fluidized bed (catalytic) permselective membrane reactors. For more general applications material scientists



Catalyst particle in a reacting medium
Where do the integration of functionalities occur?

Type A : at the catalyst level, multifunctionality : combining catalytic properties and engineered catalyst structure (bifunctional, attrition resistant, encapsulated homogeneous, thin film, dry zeolie catalysts)
Type B : at the reaction inter-phase, multifunctionality : chemical reaction and enhanced inter-phase transport
Type C : at the intra-reactor level, multifunctionality : marrying chemical reaction and intra-reactor unit processes such as heat transfer or separatons (catalytic distillation, membrane reactors...)
Type D : at the inter-reactor level, multifunctionality : chemical reaction with recirculatin solids (moving-bed or circulating fluid-bed-reactor FCC : combining two reactors operations

Fig. 5. Process intensification using multifunctional reactors [11].

must solve the problem of providing inorganic membranes of perfect integrity involving mechanical and thermal stability and which will allow large fluxes of desired species and second, chemical engineers must figure out the heat transfer problem which now threatens successful scale-up.

Finally, though multifunctional reactors are not quite new to the process industries, i.e., absorption or extraction with chemical reaction, only recently reactors incorporating several "functions" in one reactor have been formally classified as being multifunctional and the large benefits obtained in integrating progress of knowledge at different scale and time-lengths have been acknowledged by the process industries. This was illustrated by the first international symposium on multifunctional reaction in 1999 [10]. But to achieve optimal performance with multifunctional reactors, it is important to lead a scientific approach to understand where do the integration of functionalities occurs, as explained in Fig. 5 [11] in the case of a catalyst particle in a reacting medium.

However, we will mention more generally that the use of hybrid technologies encountered in a great number of multifunctional reactors is limited by the resulting problems concerning control and simulation, i.e., the interaction between simultaneous reaction and distillation introduces more complex behaviour involving the existence of multiple steady states and output multiplicities corresponding to different conversion and product selectivity, compared to conventional reactors and ordinary distillation columns. This leads to interesting challenging problems in dynamic modelling, design, operation, and strong non-linear control.

3.2.2. Process intensification using new operating modes

The intensification of processes may be obtained by new modes of production which are also based on scientific principles. Indeed new operating modes are in the laboratory and/or pilot stage: reversed flow for reaction-regeneration energy efficient coupling of endo- and exothermic reactions, countercurrent flow and induced pulsing flow in trickle beds, unsteady operations, cyclic processes, extreme conditions, pultrusion, low-frequency vibrations to improve gas–liquid contacting in bubble-columns, high temperature and high-pressure technologies, and supercritical media, and use of composite structured packings achieving low pressure drop through vertical stacking of catalyst, are now seriously considered for practical application.

3.2.3. Process intensification using microengineering and microtechnology

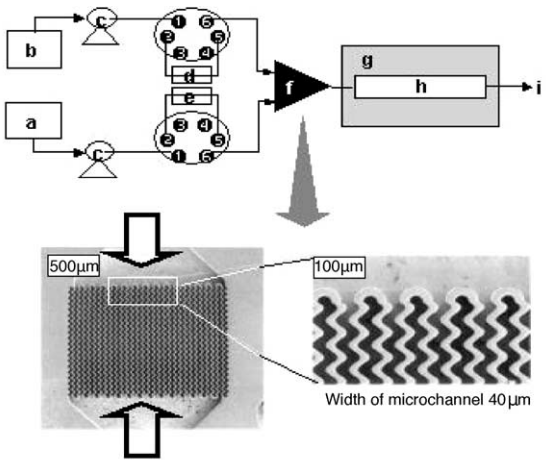
Current production modes also are and will be more and more challenged by decentralization, modularization and miniaturization. Microtechnologies recently developed, especially in Germany (i.e., IMM, Mains and Forschnungszentrum, Karlsruhe) and in USA (i.e., MIT and DuPont) lead to microreactors, micromixers, microseparators, micro-heat-exchangers, and microanalyzers, making possible accurate control of reaction conditions with respect to mixing, quenching, and temperature profile.

Miniaturization of chemical analytic devices in micro-total-analysis-system (μTAS) represents a natural extension of micro-fabrication technology to biology and chemistry with clear applications in combinatorial chemistry, high-throughput screening, and portable analytical measurement devices. Also the merging of μTAS techniques with microreaction technology promises to yield a wide range of novel devices for reaction kinetic and micromechanism studies, and on-line monitoring of production systems [12].

Microreaction technology is expected to have a number of advantages for chemical production [13,14] as the high heat and mass transfer rates possible in microfluidic systems allow reactions to be performed under more aggressive conditions with higher yields that can be achieved with conventional reactors. Also new reaction pathways considered too difficult in conventional microscopic equipment, e.g., direct fluorination of aromatic compounds, could be pursued because if the microreactor fails, the small amount of chemicals released accidentally could be easily contained. And the presence of integrated sensor and control units could allow the failed microreactor to be isolated and replaced while other parallel units continued production. Also these inherent safety characteristics could allow a production scale systems of multiple microreactors enabling a distributed point-of-use synthesis of chemicals with storage and shipping limitations, such as highly reactive and toxic intermediates (cyanides, peroxides, azides) [13].

Moreover, scale-up to production by replication of microreactors units used in the laboratory would eliminate costly redesign and pilot plant experiments, thereby shortening the development time from laboratory to commercial-scale production. This approach would be particularly advantageous for pharmaceutical and fine chemicals industries where production amounts are often less than a few metric tons per year.

Also it was proposed a new concept for high-throughput screening (HTS) experiments for rapid catalyst screening



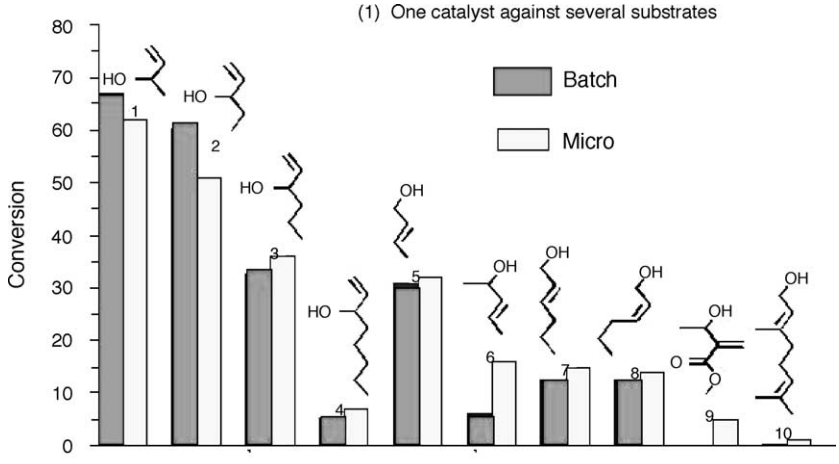
Experimental set up : a water reservior, b.n-heptane reservior, c.high pressure liquid pumps, d and e, HPLC type injection valve equipped with 200ml and 1ml loop, f.MICROMIXER. G thermoregulated bath, h. tubular stainless steel reactor (80cm, lenght, 0,4cm i.d), i outlet analytics, j SEM image of the mixing micro element showing the 2x15 interdigited microchannels (25?m width corrugated walls.

Fig. 6. The IMM micromixer for high-throughput screening [15].

based on dynamic sequential operations with a combination of pulse injections and micromachined elements [15]. The authors describe a new concept to achieve HTS of polyphasic fluid reactions for two test reactions, a liquid–liquid isomerization of allylic alcohols and a gas–liquid asymmetric hydrogenation. The principle used for the test microreactor is a combination of pulse injections of the catalyst and the substrate, a IMM static micromixer with negligible volume and residence time less than 10^{-2} s, and a tubular reactor. The two scanning electron microscopy images show the micromixer, in which 2×15 interdigitated microchannels (25 μm width) with corrugated walls are clear (Fig. 6). The pulses mix perfectly in the micromixer and the liquids or the gas–liquid mixtures thereby emulsify and behave as a reacting segment, which then travels along the tubular microreactor.

Collection at the outlet of the reactor and analysis afford the conversion and selective data. The catalyst library was then screened. The results led to the selection of the best catalyst showing activity towards a large class of allylic alcohols. Similar results which were obtained in a microreactor and in traditional well mixed batch reactor (40 cm^3) proves the validity of the concept (Fig. 7).

In term of catalyst and time consumption per test, the numerous tests for the liquid–liquid isomerization were performed twice, to test for reproducibility, using only 1 or 2 μmol of metal and over a total screening time of 1 h. The test for the gas–liquid asymmetric hydrogenation showed similar features (down to 0.2 μmol of catalyst, and 3–5 min per test). Throughput testing frequencies of more than 500 per day are achievable, albeit with computer control of the apparatus. Using these microreactors for dynamic, high-throughput screening of fluid–liquid molecular catalysis offer considerable advantages over traditional parallel batch operations: ensuring good mass and heat transport in a small vol-



Catalyst pulse 0.2 $cm^3$of a [RH] 0.004 $kmol/m^3$ solution equivalent to 80μg of Rh for each test. Catalyst RhC1^3/TPPTS/NaOH. Rh/TPPTS = 5. Selectivity . 95%.70°C. Flow rate : Aqueous phase 5 cm^3/min Organic phase 1 cm^3/min [substrate] 0.1 km ol/m^3 Residence time 100 s.

Fig. 7. Comparison with traditional equipment (batch reactor) [15].

ume, reduced sample amounts (to μg levels), a larger range of operating conditions and simpler electro-mechanical moving part.

3.3. Product design and engineering: manufacturing end-use properties: development of multidisciplinary product-oriented engineering with a special emphasis on complex fluids and solids technology

This is the answer for the nowadays ever-growing market place demand for sophisticated products combining several functions and properties: cosmetics, detergents, surfactants, bitumen, adhesives, lubricants, textiles, inks, paints, paper, rubber, plastic composites, pharmaceuticals, drugs, foods, agrochemicals, and more.

This product design and engineering (synthesis of properties), is the translation of molecular structure into macroscopic phenomenological laws in terms of state variables and in practice it mostly concerns complex media and particulate solids. Indeed complex media such as non-Newtonian liquids, gels, foams, hydrosoluble polymers, colloids, dispersions, emulsions, microemulsions, suspensions for which rheology and interfacial phenomena play a major role are often involved. Also involved are the so-called "soft solids", systems which have a detectable yield stress, such as ceramic pastes, foods or drilling muds.

Product design concerns also particulate solids encountered in 70% of the process industries. This involves the creation and the control of the particle size distribution in operations such as crystallization, precipitation, prilling, generation of aerosols and nanoparticles as well as the control of the particle morphology and the final shaping and presentation in operations such as agglomeration, calcination, compaction, encapsulation. Both types of operations need a better understanding as they control the end-use property and quality features, such as taste, feel, smell, colour, handling properties, sinterability or biocompatibility. Product design and engineering concerns also solids considered as vehicles of condensed matter from the perspective of solventless processes or non passive "intelligent solid" to accomplish intelligent functions such as controlled reactivity or programmed release of active components that may be obtained by multiple layer coatings.

The quality and properties of emulsified or past like and solid products is determined at the micro- and nano-level. Therefore to be able to design and control the product quality and make the leap from the nano-level to the process level, chemical and process engineering involved with structured material have to face many challenges in fundamentals (structure–activity relationships on molecular level, interfacial phenomena, i.e., adhesive forces, molecular modelling, i.e., equilibria, kinetics, product characterization techniques, etc.), in product design (nucleation growth, internal structure, stabilization, additives, etc.), in process integration (simulation and design tools based on population balance) and in process control (sensors and dynamic models).

For illustration, we may cite the control of the quality of microemulsions for foodstuffs containing microorganisms that could spoil and whose growth can be prevented by enclosing them in a water-in-oil emulsion of aqueous droplet size not significantly larger than 1 μm and of a narrow size distribution, which namely characterizes the product quality [16]. Such miniemulsions can only be generated in high-pressure homogenizers with a high-energy input and customized nozzle geometry. However, the droplets generated must not coalesce during emulsification which makes it necessary to find emulsifier systems which also stabilize the droplets sufficiently fast. So, in modelling the emulsification process, the overall process has to be divided into two substeps: generation of droplet by mechanical energy and stabilization of the droplets before they re-coalesce. The resulting product quality is determined not only by how well the dispersed phase has been broken up into small droplets but also by how well the equipment, process conditions and emulsifier have been matched to one another. Thus, the kinetics of the molecular process determines whether the desired end-product properties will really be achieved, even if the required droplet size had been achieved in the first substep.

Complementary in topics such as microemulsions for chemical, food and pharmaceutical industries (drug delivery systems), it should be emphasized recent investigations on monodisperse emulsion formation with micro-fabricated microchannel (MC) array, called straight through microchannel, i.e., silicon array of elongated through-holes for monodisperse emulsion droplets [17]. Such oblong straight through MC equipment allows to get monodisperse oil in water emulsion droplets with average diameter of 32.5 μm and a coefficient of variation of 1.5% verifying their excellent monodispersity (Fig. 8). Such monosized droplets in

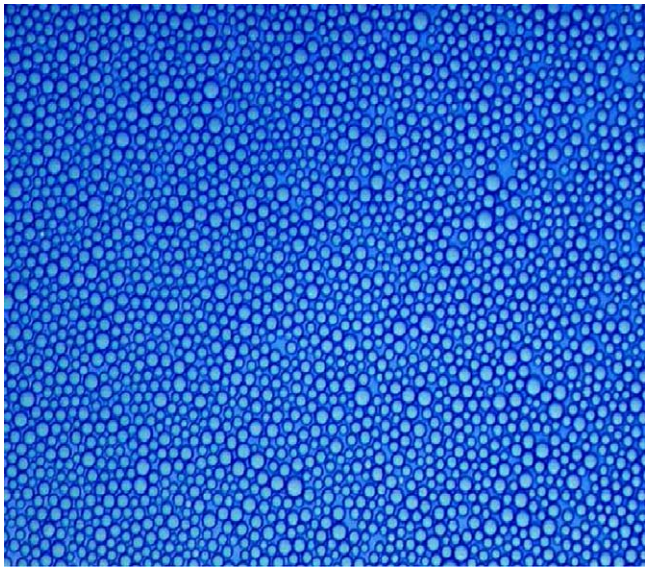


Fig. 8. Monodispersed emulsion formed with micro-fabricated microchannel array.

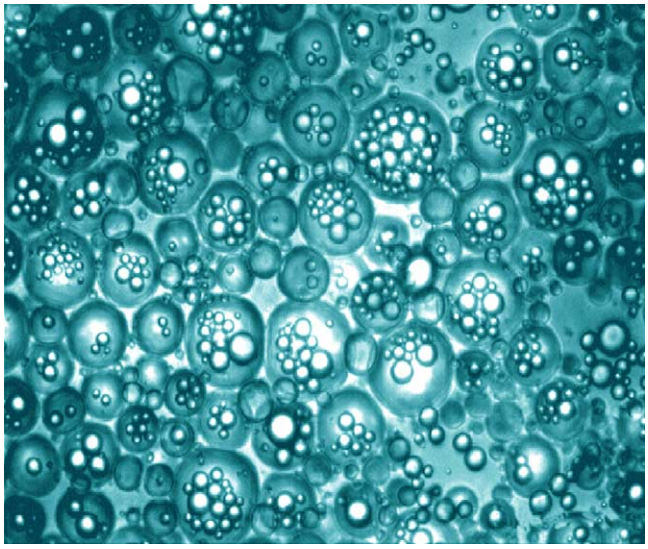


Fig. 9. Multiple W/O/W emulsion manufactured by partial-phase soluinversion technology [18].

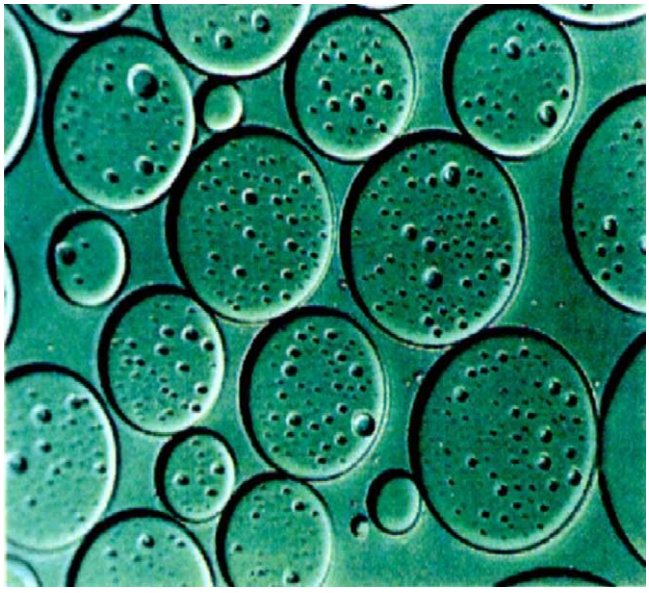


Fig. 10. Solid lipid nanoparticles.

emulsions have advantages for control of their physical and functional end-use properties, stability and application to other processings.

For topical delivery especially on the skin, novel multiple lipidic systems account for sustained release and optimized stabilization of active ingredients as well as drugs. Topical delivery for cosmetic products combine aspects of optimized skin release of actives and an optimized match to sensorial features of a product. Prominent examples for preparation of such kind are multiple emulsions of the water-in-oil in water type (W/O/W type), produced by the partial-phase soluinversion technology (PPSIT), and solid lipid nanoparticles (SLN, lipopearls) and multicompartment solid lipid nanoparticles (MSLN).

Multiple emulsion based on the PPSIT technology (Fig. 9) combine protecting and occluding effects of classical W/O emulsions and easy application feature of classical O/W formulations. Besides, the W/O/W base as such already shows excellent skin caring properties, as exemplified by improving skin's microrelief, short-, mid-, and long-term moisture holding capacity (adaptogenic moisturization) and skin firmness improvement. Such multiple emulsions are manufactured by novel one step manufacturing technology even facilitating industrial scaling up to large scale (up to 1 t batches).

Formulation of oxidation-instable ingredients such as lipoic acid and retinol are preferentially stabilized in solid lipid nanoparticles (SLN) suspensions, which can either coat the instable materials as solid shell or even can entrap additional solving oil compartments to be detected active (Fig. 10). SLN particles can be manufactured based on proloxamer derivatives as well as non-ethoxylated lipids, such as compritol or dynasan. High-pressure homogenization reveals also ultra narrow particle size distribution in the nanometer range and an excellent stabilization of lipophilic ingredients such as Ubiquinone Q10 and Vitamin E and derivatives. Due to the solid character of this carrier, active ingredients can either be protected against oxidation and hydrolysis.

For other illustration we may cite the control of the shape and size of crystals in an industrial crystallization process. It has been shown that much improved process control, both in terms of crystal purity and a defined size distribution could result by detailed computer studies of the crystallization processus which can be remarkably changed by the presence of small traces of foreign substances such as unwanted by-products in the feed solution [19,20].

Indeed in order to understand the mechanisms causing these changes in crystals size and shape so that it is possible to utilize them in a controlled manner, one must explain the structure–activity relationships on a molecular level. And with computer simulations, diagrams of the molecular structure of the most important crystal surfaces can be generated from X-ray crystal structure data.

Also in the same computer simulations, contaminant molecules or molecules with an expected beneficial effect on crystallization processus can be placed on each crystal surface and their adsorption energy calculated. If the hypothesis is that the growth rate of surface decreases with increasing adsorption energy, and by comparing relative adsorption energies the modified crystal shape to be expected can be predicted. This was illustrated with the results of crystallization from a feed ammonium sulphate solution containing dye amaranth. It was shown that the molecule amaranth is adsorbed onto 0 0 1 surface of ammonium sulphate crystals with the highest adsorption energy in comparison with the other crystal surface. And according to the calculations, the somewhat block-shaped crystal produced in the pure system becomes a flat shaped crystal having a large 0 0 1 surface area, which was experimentally verified (Fig. 11 [19]). Comparable prediction were obtained in the case of a feed ammonium sulphate solutions containing 50 ppm Al^{3+} [20].

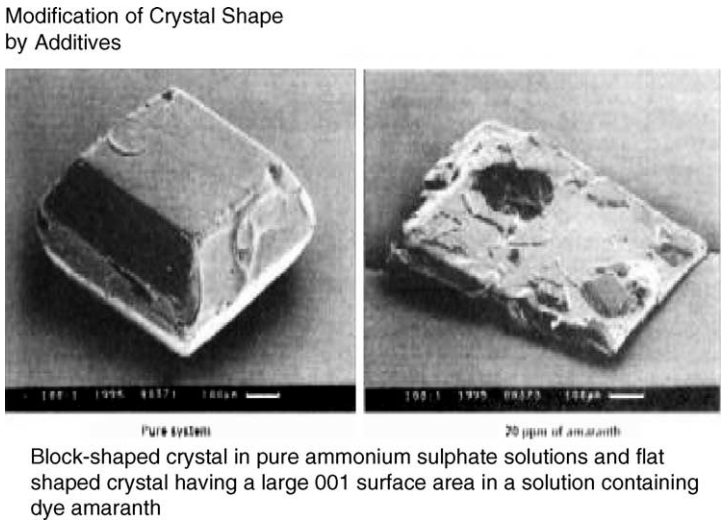


Fig. 11. Product-oriented engineering: controlled crystallization process [19].

So many quality features can only be designed in a targeted way if the molecular processus are understood at this level. And as shown by this example the analysis—both by theoretical and by experimental means—must be carried out down to the molecular level to obtain results of real value for understanding the relationship between a certain set of product qualities and the physical product state: indeed it is well known that two pain killing tablets may have the same chemical composition, but different routes of production may lead to different crystallinity and porosity profiles and therefore to totally different dissolution and solubility properties, namely different bioavailabilities.

New approaches offer now the possibility of accurate predicting the effect of solvents or impurities on crystal shape [21] and recent models recognize the significance of interfacial phenomena in crystal shape modelling, and lead the way for future developments, such as new simulation and/or group contribution methods for interfacial free energy production.

Another illustration of this multilevel research effort in crystallization has been proposed in the area of electrical engineering: microelectronics. It concerns correlations between operating conditions and microstructure of low pressure chemical vapor deposit (LPCVD) silicon based films prepared from silane, SiH_4. The main aim is the development of a rigorous simulation model for the interpretation of the layer growth experimental data by taking into account both the mass transfer resistance at boundary layer and the solid layer growth kinetic expression [22]. The four blocks of accessible knowledge for a LPCVD process are presented in Fig. 12. Specialists in material sciences may relate microstructure and properties of thin films while specialists in chemical engineering can correlate and simulate the macroscopic operating conditions of a CVD reactor and the local conditions inside the reactor. It is clear that any relationship established between blocks 2 and 3 affords an intrinsic property of the system analyzed which is independent of the equipment used and which allows to treat the complete design or optimization problems completely by theoretical methods. For example it should be possible to treat, first, the kinetic questions such as to find sets of conditions producing the desired thickness with the desired degree of uniformity. In a second step, one could solve the questions of material structure (amorphous, partially or totally crystallized silicon) end-use properties.

The thin CVD layers were produced in the so-called hot wall tubular reactor (2 m length, 2.2 m in diameter). Numerous characterization methods have been used by the authors to study the layers microstructural evolutions in function of their elaboration conditions. The local state of the reactor was simulated by a model CVD including the gaseous flow hydrodynamic and the mass transfer and chemical reaction parameters both in heterogeneous phase (gas–solid interface) and in homogeneous phase (solid phase). Mechanism existing at surface substrate and nucleation and crystal growth

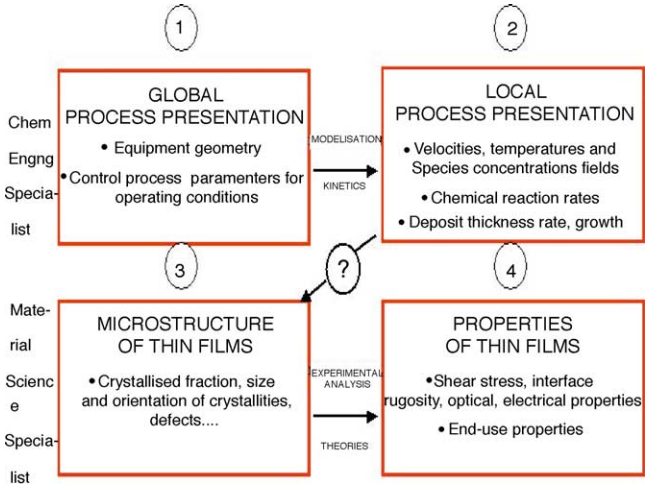


Fig. 12. The four blocks of an accessible knowledge for a CVD process: the aim, relation between 2 and 3 intrinsic property of the system (independent of the reactor geometry) [22].

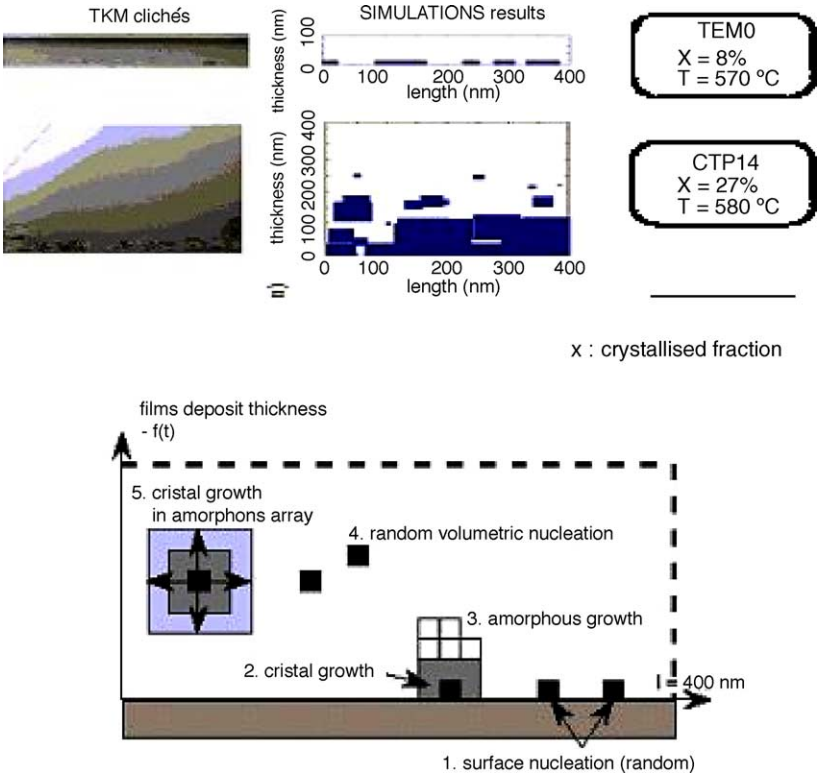


Fig. 13. Microstructure of silicon LPCVD films from silane [22].

phenomena together with the dynamic characteristic of the microstructure formation were modellized with simple geometric and statistic approach based on concepts of the mechanics of continuous media (instead of using molecular dynamic models requiring too much computer power). The final model that necessitates the local conditions of elaborations at surface substrate and nucleation and crystal growth laws, is able to calculate the thickness, the fraction of crystallized silicon and the space distribution of silicon crystallites in the deposit layer. So with such a model it is possible to predict the microstructure of LPCVD films from operating conditions such as gas phase components, temperature, pressure and initial substrate nature or surface state. A comparison between the transmission electronic microscopy micrographies of the experimental silicon films from pure silane for different temperatures and deposit durations and the simulation of the model has shown a good approximation for the crystallized fraction x_c and the space distribution of crystallites (Fig. 13). The results also emphasize the dynamic characteristics of film microstructure. Similar results were obtained for complex SIPOS SiO_x deposit elaborated from silane and nitrogen protoxide and in situ boron silicon deposits from silane and boron trichloride [23]. It is interesting to note that the quality of such simulation results demonstrates the validity of the approach proposed and suggests the way now opened to develop a complete product design and engineering or engineering of materials elaboration, able to predict the kinetic and structural characteristics of LPCVD films by numerical simulation.

Finally, much progress have been realized these last few years for the product design and the control of the process using the scientific methods of chemical engineering. Thermodynamic equilibrium states are examined, transport processes and kinetics are analyzed separately and these are linked again by means of models with or without the help of molecular simulation and finally with the help of computer tools for simulation, modellization and extrapolation at different scales for the whole supply chain (BASF, Unilever, Degussa, Astra Zeneca, Nestlé, etc.).

But how can operations be scaled up from laboratory to plant? Will the same product be obtained and will its properties be preserved? What is the role of equipment design in determining product properties? How can the optimal interactions between product and process design be explored? How can we validate and test the desired functional properties (i.e., controlled drug release, enhanced bituminous or textile behaviour, skin improvements in using cosmetic creams, etc.) of the product in use? Indeed as underlined many times, the control of end-use properties is a key issue for which general scale-up rules are still lacking. This requires for chemical engineering specialists and their systemic approach a close cooperation with specialists in physical chemistry, biology, mechanics and mathematics to develop this new “systemic” physical chemistry and biology where qualitative explanation will be translated with the help of fine modellization into formal laws for process development. This leads to the fourth main objective.

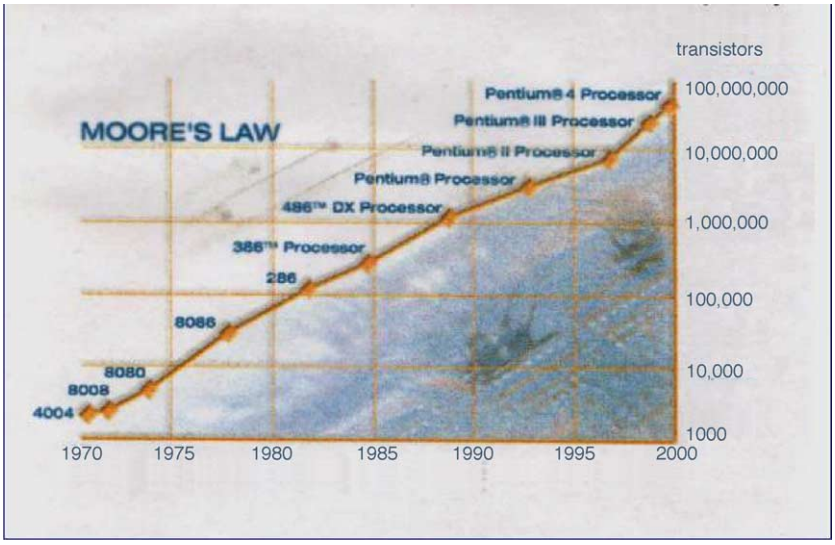


Fig. 14. Moore's law number of transistors on a silicon would double every 2 years.

3.4. Implement multiscale and multidisciplinary computational chemical engineering modelling and simulation to real-life situations: from the molecule to the overall complex production scale into the entire production site

We have emphasized the necessary multidisciplinary and multiscale integrated approach applied to the triplet processus–product–process to scale from the nano- and microscales of end-use properties of the product to the mesoscale of the equipment manufacturing the product.

Computers have opened the way for chemical and process engineering in the modelling of molecular and physical properties on the nano- and microscopic scales. For the molecular modelling, application of the principles of statistical molecular mechanics computational techniques (Monte Carlo and molecular dynamics) and quantum mechanics constitute an area for the problem-oriented approach of chemical and process engineering. Indeed molecular modelling starts from a consideration of microscopic structure and molecular interactions in a material system and derives thermodynamic, transport, rheological, mechanical, electrical, electronic or other properties through rigorous deductive reasoning bases on the principles of quantum and statistical mechanics. Compared to more phenomenological approaches (e.g. correlations of the group contribution type) it offers the advantages of greater, generality and reliability.

It is clearly impossible to cover all directions of present-day molecular modelling researches involved in a wide spectrum of problems in the chemical and material sciences. Vapor–liquid equilibria, vapor–liquid–liquid equilibria, liquid–solid equilibria, supercritical solution properties; amphiphiles; polymers at interfaces; adsorption on surfaces and influence of impurities; microporous materials or ceramics structures; transport properties such as viscosity, diffusivity, thermal conductivity can be calculated today by molecular modelling based on information from thermodynamic, kinetic and rheological data banks.

There is no doubt that molecular modelling is now playing an increasingly important role in future chemical and process engineering research and practice [24]. Recent advances in the fundamental molecular sciences and in computer hardware and numerical algorithms have greatly accelerated its development (see Figs. 14 and 15, where it is shown that the number of transistors on a sliver of silicon would double every 2 years).

And through the interplay of molecular theory, simulation, and experimental measurements evolves a better quantitative understanding of structure–property relations, which, when coupled to macroscopic chemical engineering science, can form a basis for new materials and process design.

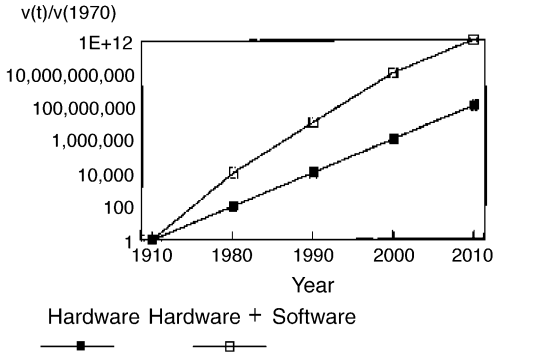


Fig. 15. Computing speed acceleration.

Furthermore, turning to the macroscopic scale, dynamic process modelling and process synthesis are being also increasingly developed. Indeed one must remember the targeted products in question are generally not mass-produced products but ones which are produced in small batches and just in time for delivery to the customer whose needs are constantly changing and evolving. And to be competitive under these conditions, it is particularly important to analyze and optimize the supply chains for which we are interested in the time that individual process steps take, and these have to be simulated and evaluated also in terms of costs. But in chemical and related processes, the location of a particular component in the supply chain at a given time is not well defined. Indeed a batch can be found in a stirred tank, a filter, a dryer, a pump, a mill and a storage container simultaneously. New event-driven simulation tools help solve these problems by simulating both material flows and states within the individual pieces of equipment. This dynamic simulation may enable to see in a matter of seconds whether bottle-necks may occur in the plant over the course of days, months or years. These can be eliminated by using additional pieces of equipment or by making additional resources available such as energy or manpower. The event-drive simulation also shows which alternative plant and storage strategies provide the greatest cost–benefit [19].

Moreover, the ability to use a series of coherent tools (i.e. software) during the process life cycle is now well established: integration and opening of modelling and event-driven simulations environments as answers to the current demand for diverse and more complex models in process engineering is taking today a more and more important place, see, for example the Computer Aided Process Engineering European Brite Euram program CAPE-OPEN. The aim is to promote the adoption of a standard of communication between simulation systems at any time and length scale level (thermodynamic, unit operations, numerical utilities for dynamic, static, batch simulations, fluid dynamics, process synthesis, energetics integration, control process) in order to simulate processes allowing the customers to integrate the informations from any simulator onto another one. For the future, the global CAPE-OPEN (GCO) project is expanding and developing interface specification standards to insure interoperability of CAPE software components. And a standardization body (CAPE-OPEN Laboratories Network, CO-LaN) has been established to maintain and disseminate the software standards in the CAPE domain that have been developed in the international projects CAPE-OPEN and Global CAPE-OPEN. The CO-LaN ensure that software tools used by the process industries reach a level of interoperability that will help ensure sustained growth and competitiveness [25,26].

4. Conclusions: chemical and process engineering mainly concerned by the production of new materials

The production of new materials to satisfy consumer needs and market trends requires the development of chemical and process engineering involving an integrated multidisciplinary and multiscale approach from the molecular-scale and material surface-scale-up to the scale of entire production site. This multidisciplinary and multiscale approach (3P engineering, molecular processes, product, process engineering) is necessary for the understanding and modelling of the complex, simultaneous and often coupled phenomena and processes taking place on the different scales of the chemical supply chain. It is clear that this integrated approach is possible today, thanks to considerable progress in the use of molecular modelling [27], scientific instrumentation [28–30], and powerful computational tools and capabilities, as illustrated with the determination of physical and (bio)chemical parameters necessary for the modelling at different time and length scales.

Therefore for the production of classical or new materials it seems clear that the future of chemical and process engineering is heading in four directions: (1) increase selectivity and productivity by a total multiscale control of the processes, a good illustration is the nanostructural tailoring

Table 1

Trends in European chemical engineering	1999, Montpellier, ECCE2 (%)	2001, Nürnberg, ECCE3 (%)	2002, Praha, CHISA (%)	2003, Granada, ECCE4 (%)	2003, Berlin, CHEMREACTOR 16 (%)
1 Increase productivity and selectivity: through multiscale control of the process, molecular information engineering and nanotailoring of porous and crystalline materials	33	15	22	24	33
2 Process intensification: multifunctional reactors, new operating modes, microengineering, microtechnology	25	39	30	22	31
3 Formulation product design and engineering: emphasis complex fluids emulsion solids technology	22	25	14	18	5
4 Multiscale and multidisciplinary: computational chemical engineering, modellization and simulation and optimization–control–safety	20	21	34	36	31

of required materials with controlled structure or in obtaining physico-chemical and mathematical bases of processes occurring on catalyst surface, (2) process intensification by the design of novel process and equipment based on scientific principles and new production operating methods, examples concern new types of chemical processes and reactors, (3) synthesize structured products combining several functions and properties required by the customer with special emphasis on complex fluid and (soft) solids technology, (4) implement multiscale and multidisciplinary computational and simulation to real-life situations with an emphasis on the understanding of the physics, chemistry and biology of the interactions involving control and safety considerations.

Table 1 shows the distribution of the contents of the lectures and posters presented during the recent European congress in chemical engineering including the XVI International Conference on Chemical Reactors in Berlin, 1–5 December 2003. It may be seen the increasing part of the academic and industrial investigations in modellization, simulation, optimization, control and safety.

And in the future within the frame of its four precited objectives, chemical and process engineering will be more and more concerned and involved especially with the application of *life cycle assessment* to the triplet "processus–product–process", i.e., application not only to product design and engineering and its use but also to the plant and equipment together with the associated services.

Thus, this multidisciplinary and multiscale integrated approach will also be of great help, in responding to the increasing environmental, societal and economic requirements and to the transition towards sustainability.

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