Nature Inspired Chemical Engineering

Inaugural lecture

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Mijnheer de Rector Magnificus,
leden van het College van Bestuur,
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van de universitaire gemeenschap.
Zeer gewaardeerde toehoorders.
Dames en heren,

I will present my "oration", my inaugural lecture, in English rather than in Dutch, mainly as a courtesy to several friends and colleagues who have come from abroad.

Research Philosophy

Friends know how I like to travel. Exposure to different environments refreshes the mind, and stimulates awareness. It provides possible answers, but also brings up many new surprises and questions that require further investigation, which is what science is all about; our understanding of the world is minute, and there is, in my opinion, no "end of science" in view. This is, therefore, an exciting time to be a scientific researcher. It is also a lot of fun. One is free. Like in art, there is the comfort *and* the burden of history, the possibility to learn from past work, yet also to challenge it, to innovate, and to create. Research is an exciting adventure, with some planning, some maps, yet also a lot of uncharted territory. As Albert Szent-Györgyi, Noble laureate and discoverer of vitamin C said: *Discovery consists of seeing what everybody has seen and thinking what nobody has thought.* Many are the opportunities on this fantastic voyage to follow badly marked roads or even allow a random walk along beautiful sites that may help to provide one more answer, before hurrying on to a new adventure, with a little more guidance every time. Most problems still remain to be solved. Helped by a simultaneously developing mathematical language, our understanding of nature does not go beyond some of the fundamentals of equilibrium thermodynamics, physics, hereby also chemistry, and, just starting, a little bit of biology, while we have at most an incomplete, phenomenological, empirical overall description of almost anything else. A multi-disciplinary approach, combining knowledge and experience from a variety of disciplines, is essential in identifying and solving new problems. Because so many problems still remain to be solved, choices are virtually unlimited and being an academic scientist is a fascinating profession. However, the scientific profession is entirely personality driven, for it is personality that makes one choose to follow a certain road, and to study one particular problem rather than another. While I enjoy taking occasional random excursions and find them useful to take a fresh look at things, in essence I do not take scientific research as a random walk. Most of all, I cannot do but follow the passions that characterize my research and are best expressed *via* the title of my inaugural address: *Nature Inspired Chemical Engineering*.

In my oration, I will mainly talk about my passion: what I mean with "nature inspired," why I focus on this topic, how my interests in this area are developing. I will draw a sketch, not present a completed painting. I will give a few examples from my current research, and questions I may work on in the future. Many postulates. A few answers, but many questions – this is an *inaugural* speech after all, a beginning. Five years ago, I did not know I would ever work in Holland, let alone be standing here in this function today. I do not know where the present adventures will take me in five more years, what I will work on then. That too, is part of the excitement of my profession. It is common to scientific researchers and artists, and I consider it the excitement of life itself, for what pleasure is there in trying to live the future already, when there is the present, and the future will come, whatever we do?

Nature Inspired Chemical Engineering is first about chemical engineering. This is not only an exciting time to be a scientist, but also one to be an engineer, and in particular a chemical engineer. Chemistry is the science of change or transformation and, hence, is the interface between physics and biology and materials. Engineering is designing and making things work, and is therefore the interface between science and society. *Engineering is a humanity*, as John Prausnitz, Professor in thermodynamics at the University of California, Berkeley, points out. We cannot ignore our environment. The real scientific and engineering challenges are related to our socio-economic and ecological environment. Here, the challenges are enormous. For centuries, progress has been mostly associated to an increasing *domination* over nature, *i.e.*, applying our knowledge to use or rather submit nature. Engineering became artificial. This seems to be a human characteristic, and especially a Western one: using limited resources, taking without returning, wasteful overproduction and overconsumption. According to George Bernard Shaw, *The reasonable man looks at the world around him and tries to adapt himself to it. The unreasonable man tries to adapt the world around him to himself. All progress, therefore, is dependent on the unreasonable man.* We have changed nature, by trying to control our environment and fit it to our demanding lifestyle. Engineering has mostly been viewed as a victory, an expression of superiority. Marinetti's futurist movement in art expresses this victory of the machine in an ecstatic way. This ecstasy was revolutionary at its time, and aimed at liberation from a conservative past. It also led to fascism, however: moderation is a virtue. Times have changed, and it is becoming clear that domination comes at a cost. Nature is not our student; we are nature's student, and we have not understood nature yet. Laozi, the philosopher of the *Dao de Jing*, asks: *What is stronger, water or stone?* Water flows softly around the hard stone in the river; yet, in the end, the persistence of pliant water erodes the stone until it disappears, and, therefore, water is stronger. You cannot force things. Not accounting for that is potentially extremely dangerous, even if nature at least appears to be robust, able to buffer imposed changes. I would not bet on this. A first step is mending problems, such as pollution, like a disease, but this is insufficient. It is slowly becoming apparent that we really need to live in more harmony with nature. There is a Native American saying: *We do not inherit the world from our ancestors; we borrow it from our children.* Observing changes in nature is hard. One reason is that the time constants characterizing natural dynamics are typically much longer than the ones characterizing our technological "progress". Our own nature has not evolved at the same pace; we are becoming the victims of our own so-called progress. Population is exploding. Our economic systems are only slowly starting to include the price of waste. Luckily, our instinct of survival has recently led us to slowly start to act more responsibly. This permeates through politics, economics and business, as it is becoming clear that *sustainability* is key. There is a slow shift to alternative fuels, biodegradable and biocompatible materials. This is a necessity; it is an irrelevant chicken-and-egg problem whether this tendency is politically, public relations or economically imposed. More important is that societal awareness grows. The challenge is enormous, but I am optimistic. Progress in the 21st century means building a sustainable future. It is ironic that this sustainability was built in, in the most so-called "primitive" communities, be it Aboriginal or Native American, or the most ancient Asian philosophies, while so-called "advanced" Western and now global communities, have mostly neglected this issue.

I embrace a call for chemical engineering research not to be process-driven or even product-driven, as now the most urgent issue is *society-driven, sustainable progress,* with accordingly developed processes and products. This imposes new challenges for science and engineering, and, thus, fundamentally for academia. Chemical engineering plays a very important role, for the aforementioned reasons, the impact of its products and processes being so significant. It is therefore also a great candidate for change. Debates have been going on with some people claiming that chemical engineering is loosing its identity, because it is broadening so much in scope. Chemical engineering is only a century old as a discipline, and I therefore argue that it merely develops, like a person growing up and becoming more mature with age and experience. It has never been as exciting, because of its relevance and multi-disciplinarity: it is the glue between physics, materials and biology, and between science and society, and the present scientific and societal developments are such that this glue can function as never before. The role and responsibility of chemical engineering has shifted from maximum production to most efficient production, and to generating alternative, biocompatible products. I am glad that this university is actively participating in this evolution, with good connections between industry and academia, and, hopefully, a continuing support for new creative ideas that link the fundamental with the applied.

It is noteworthy that nature itself is full of internal rivalries, of a "survival of the fittest". I like to see it as a dynamic system, most likely a chaotic one, with sudden changes, not necessarily continuous but spotted with discontinuities. *Discontinuities rule* – this is, in my opinion, together with the birth of molecular life sciences, *the* major realization of 20th Century science. A first realization was that light consists of quanta, a second that sudden events can change everything fundamentally, together with long-range, mostly persistent, correlations in space and time. Subtle differences that continuously act on a system can trigger a complete change in the end. This common notion about the weather, the stock market and politics is the essence of respectively multifractals and chaos theory, and, I believe, of life itself. My mentor, Benoit Mandelbrot, whom I will come back to later, played a significant role in revealing this fact that is popularly associated to fractal geometry, but quite different and more general.

This brings me back to a particular excitement of mine – learning from Nature. If we are to aim for sustainable progress, in harmony with nature, couldn't we learn from nature itself – nature as chemical engineer? As I will illustrate, nature is full of self-assembled, self-organized patterns of interest to the chemical engineer, and, while nature is constantly changing, optimization has had its course over the millennia. I am happy and honored to give my oration on the same day as Professor de Swaan Arons, as I see conceptual fundamental advances in thermodynamics, and in particular in non-equilibrium thermodynamics, as the origin of the potentially most significant progress in, *e.g.*, chemical reaction engineering or biotechnology. Important related research through new analytical and computational work in physical chemistry, in particular in the statistical mechanics of molecular and mesoscopic systems, should further catalyze chemical engineering progress.

Research Topics

I will now come to a more specific scientific discussion of some of the concrete research I am performing and planning to perform, all around the central philosophy of *Nature Inspired Chemical Engineering*, although I reiterate that research is a dynamic, evolving process.

Precision Technology by Rational Design

Society is our starting point, our motivation. Major challenges are obtaining higher material and energy efficiencies, and a higher selectivity toward desirable products in chemical processes, as well as new functional and smart materials, in correspondence with our aim to be in a sustainable balance with our environment.

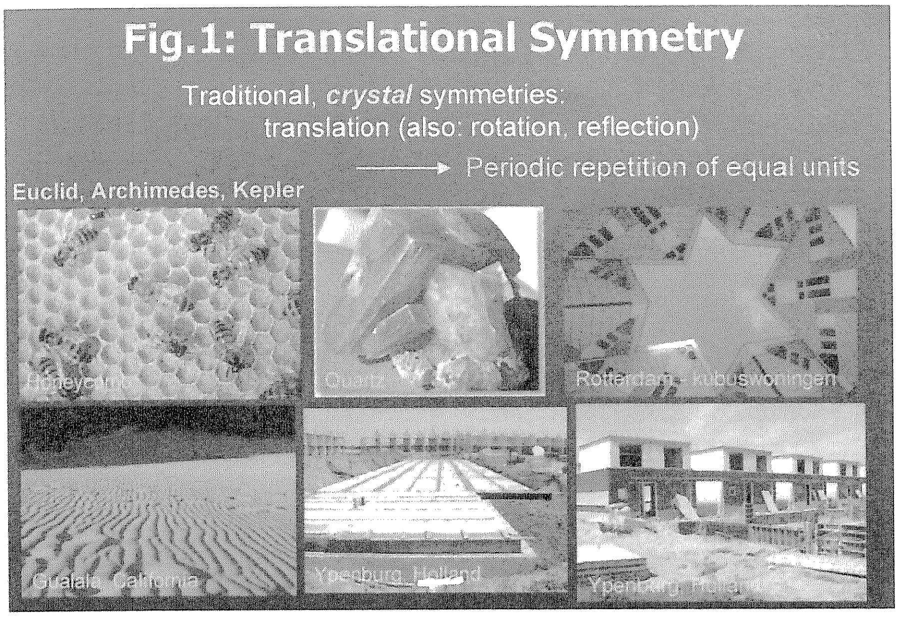
These demands require precision technology, a careful rational design of products and processes at *all* scales. My main proposition is to learn from the efficient and often elegant patterns that occur in Nature, both spatial – from molecular to macroscopic scales – and temporal – at all time scales.

Precision technology in chemical engineering embodies the control over interrelated chemical and geometric factors. Chemical specificity is required to obtain the desired highly selective chemical functionality; an example is the chemical nature of the active sites in catalysis. Geometric control should occur at *two* levels. A first is that of the building blocks, say the molecular architecture: This is the one we are most familiar with in chemistry. Examples are enzyme structure (which is related to its function, according to a lock-and-key principle), structured polymers, and nanomaterials, such as nanotubes, clusters and nanocrystals. Enormous progress has recently been made in this area. A second level, however, is the supra-molecular architecture, with which I mean the patterns that link the building blocks with the macroscopic world, essential to nature *and* to the engineer. These architectures involve hierarchically structured materials, products and processes. It is on these architectures that I mainly focus, because at least as much progress is necessary in this area to make the building blocks useful. The question then is: *How should we assemble the building blocks?*

Symmetrical Architectures

One attractive answer is putting the blocks together in an as simple as possible way, in order to obtain a maximum effect with a minimum effort. Imposing some type of *symmetry*, which in general means invariance under a transformation, would simplify the architecture significantly and hereby simplify modeling, construction and control over materials, products and processes.

The best-known symmetries are *crystal symmetries*, and in particular translations, where a displacement of the structure leads to an identical structure (Figure 1). Crystals like quartz consist of identical units that are repeated in all directions, rotated, reflected and/or displaced. These symmetries are present in quite a few patterns in nature, such as in honeycombs and waves on water, sandy beaches or dunes, and are ubiquitous in daily life, in particular in architecture.



Holland is a perfect example of this: there are the interesting arrays of "kubuswoningen" (cubic buildings) in Rotterdam, or the not-so-interesting developments in Ypenburg, a suburb of The Hague, where the house number is essential to find your way back to your own house. The overwhelming presence of these crystal symmetries in our construction, but also in our thinking, is linked to our acquaintance with Euclidean geometry from kindergarten on. Almost everything is related to straight lines, planes, regular polygons and polyhedra: Euclid, Archimedes and Kepler rule.

Another class of symmetries, however, is as ubiquitous, namely the *fractal symmetries* of self-similarity and self-affinity, where part of the object looks similar to a larger part, *i.e.*, invariant under magnification, at least within a finite range. It is only since Mandelbrot's vision that it has become so obvious that we are literally surrounded by everyday objects and phenomena in nature that contain this fractal symmetry, without crystal symmetries. Examples are shown in Figure 2: the respiratory network of the lungs, the vascular network, trees, including their branches and their roots, turbulence – beautifully visible in Leonardo da Vinci's drawing *The Deluge*, a satellite picture of the Alps that looks remarkably similar to dendrites growing like bushes along the face of a rock. The Menger sponge in the corner is a mathematical fractal, one that can be constructed to have infinite self-similarity. Just like no line can be perfectly straight, no object can be perfectly fractal; this does not matter, because what counts is the structure within the range of scales that interests us.

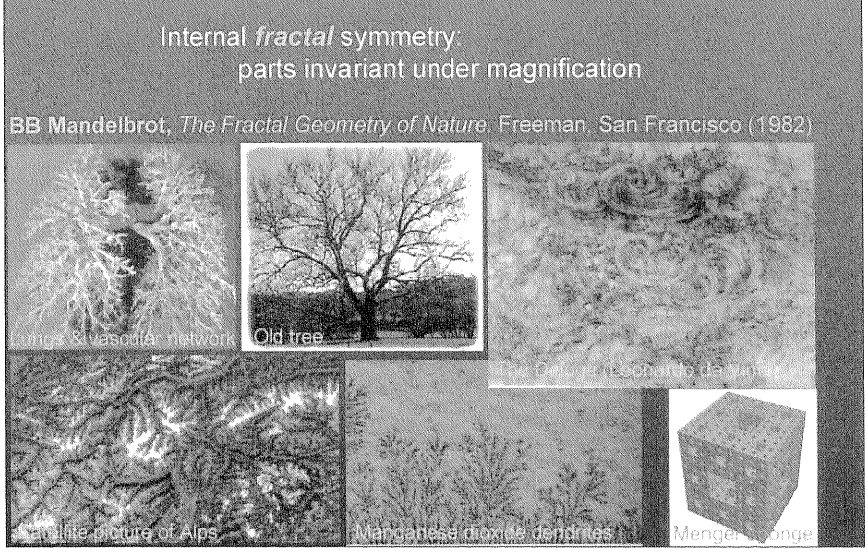
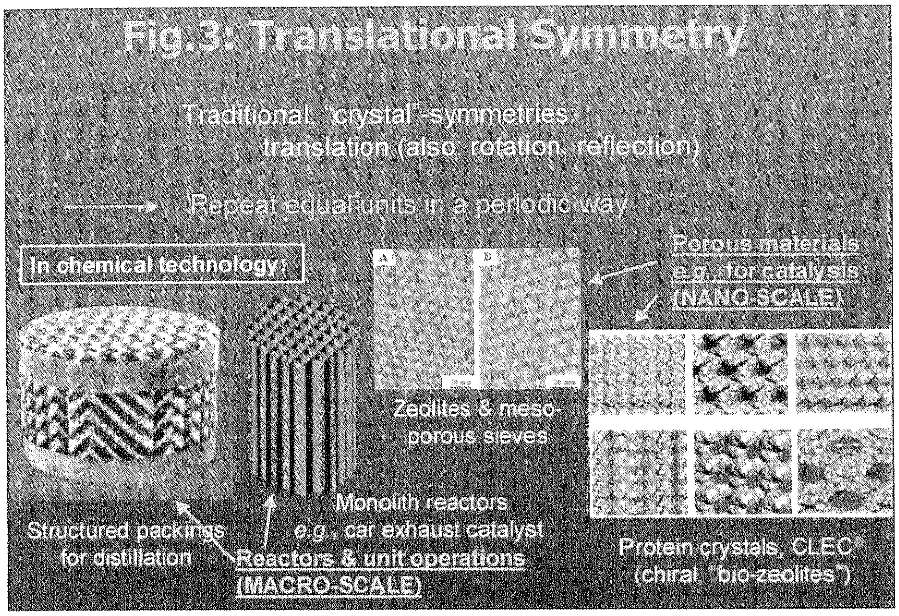


Fig.2: Self-similarity & Self-affinity

Let us move back to chemical technology and the use of the first of the symmetries – translational or crystal symmetries. A lot of research is going in this field, including by some expert groups in Delft (Figure 3). Nano-structured porous solids are being synthesized to aim for better control and higher efficiency, as the desired specific physico-chemical function can be closely related to the controlled molecular or nano-structure. A nanometer is a billionth of a meter, a millionth of a millimeter, and the size of one large or a few small molecules. Examples are zeolites, mesoporous sieves and protein crystals. In process engineering, structured reactors and separators are constructed to replace random structures, and with the same objectives of a better control, higher efficiency and easy scale-up. Monolith reactors, such as the one cleaning the exhaust of a car, as well as structured distillation packings are two examples.

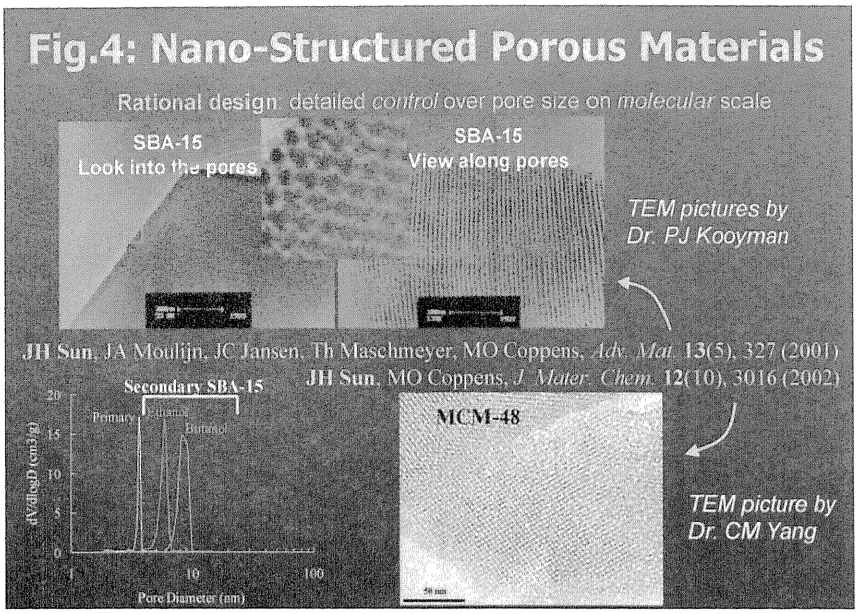
I will now discuss the nature-inspired chemical engineering approach that I propose to arrive at new materials and processes. For catalysis, *e.g.*, the combination of both catalysts (as materials) and reactors (as processes) would create an efficient, patterned reaction environment at all scales.



Nature-Inspired Porous Materials

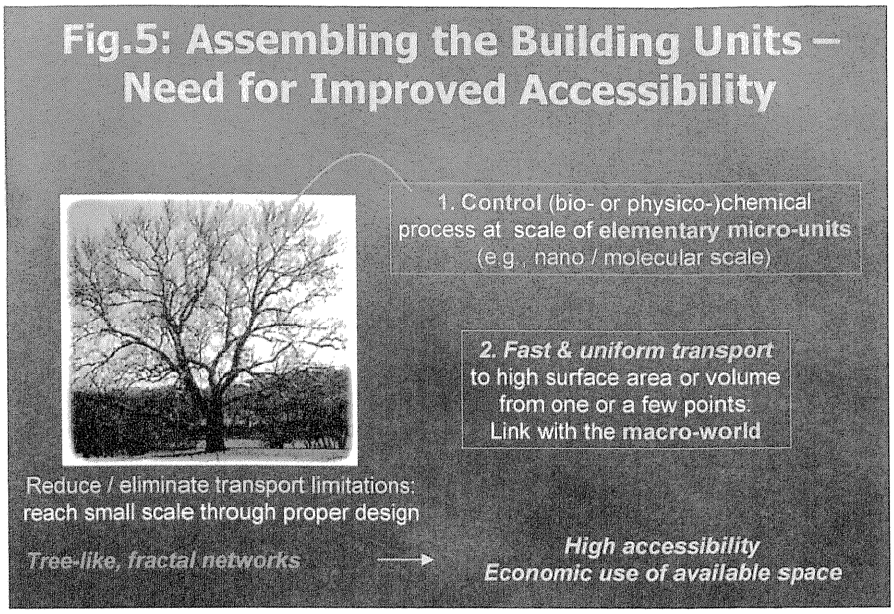
A first step is a continuation of the improvement of control at the nano-scale. In the present context, I mainly consider these materials as the building blocks I referred to earlier. Shown in Figure 4 are two materials synthesized by my former postdoc, Dr. Jihong Sun: the first, SBA-15, has an ordered hexagonal array of pores the size of which can be controlled in the nanometer range by changing the synthesis conditions; the second, MCM-48, has an ordered cubic three-dimensional pore network. By a novel technique, a high degree of stability and order could be obtained. My collaborators, mentioned here, are gratefully acknowledged. Our objective, however, is to assemble such nano-structured building units in a preferred way.

This is necessary, *e.g.*, for heterogeneous catalysis using high-area porous catalysts, where a high accessibility of the active reaction sites to the molecules that diffuse through the pores is essential. Nature solves such transportation problems in a very elegant way (Figure 5).



Observing a tree, *e.g.*, control of the essential processes at the scale of the elementary micro-units, say the leaves, is realized by the specific structure of the leaves, yet the tree can only function well if nutrients can access the leaves easily. The same holds for the lung, and easy transport of oxygen and carbon dioxide between alveoli and trachea. Fast and uniform transport to the micro-units or cells, covering a high surface area or volume, from one or a few points (say the stem) enables the efficient use of the micro-units. Transport limitations can thus be reduced or eliminated by tree-like fractal networks.

Hierarchical structures in nature abound, for easy transport of molecules and energy, but also for mechanical strength and flexibility, while making efficient use of available materials. Examples are silk, bone or tendon. These natural materials are extremely efficient and multi-functional, thanks to their composite, hierarchical structure.



How can we now synthesize hierarchical materials, such as porous materials for catalysis, which would combine the efficiency of the building units with easy molecular transport? This is an important effort in most recent years. We have come up with a method that is based upon the self-assembly of primary structured porous units around templates that are, themselves, self-assembled micelles of surfactant molecules (somewhat similar to soap).

The particles can be cross-linked by heating, form a network, while preserving the controlled nano-structure of the primary particles; removal of the surfactants leads to a hierarchical or multi-structured porous material (Figure 6). Figure 7 shows what we are doing in practice. The primary material is grainy, but the final material is more continuous by cross-linking. Using transmission electron microscopy, magnification of a small area shows the large mesopores, here 40 nm in size, and even further magnification shows the equally sized small mesopores, 3 nm in diameter. By changing the synthesis conditions, it is possible to preserve the small pore size while independently modifying the large pore size. Such control is very useful for optimization of the structure for a particular application.

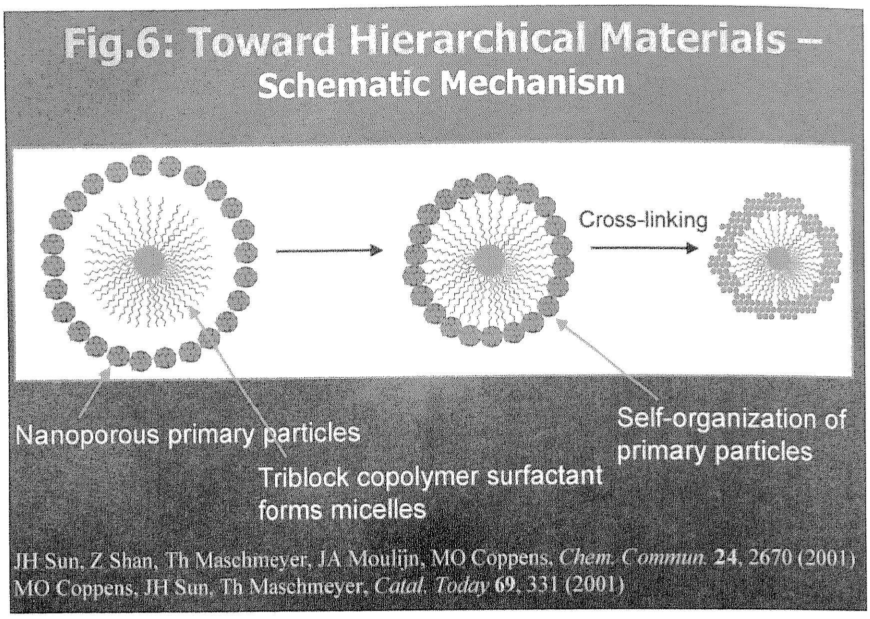


Fig.6: Toward Hierarchical Materials –
Schematic Mechanism
Cross-linking
Nanoporous primary particles
Triblock copolymer surfactant forms micelles
Self-organization of primary particles
JH Sun, Z Shan, Th Maschmeyer, JA Moulijn, MO Coppens, Chem. Commun. 24, 2670 (2001)
MO Coppens, JH Sun, Th Maschmeyer, Catal. Today 69, 331 (2001)

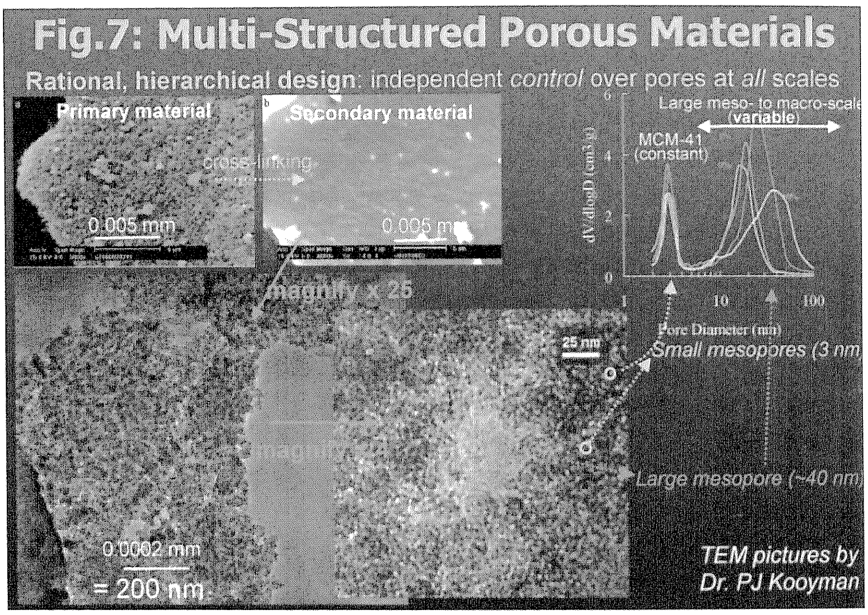


Fig.7: Multi-Structured Porous Materials
Rational, hierarchical design: independent control over pores at all scales
Primary material
Secondary material
cross-linking
0.005 mm
0.005 mm
magnify x 25
Large meso- to macro-scale (variable)
MCM-41 (constant)
dV/dlogD (cm3/g)
Pore Diameter (nm)
Small mesopores (3 nm)
25 nm
Large mesopore (~40 nm)
0.0002 mm
= 200 nm
TEM pictures by Dr. PJ Kooyman

This is merely a modest beginning, however. An improved 3D control, both textural and chemical is desired for a variety of applications. First, in chemical reaction engineering, integrated with an optimized structured reactor, applications necessitate an improved chemical and mechanical stability. An example I am recently working on with Akzo Nobel Catalysts is the synthesis of catalysts that are based upon zeolite nanocrystals, for catalytic cracking of heavy oil. Other applications include electrochemistry, electronics, optics, and life science and technology, which require a non-trivial extension of these methods to organics, proteins or composites. Most challenging is the physical chemistry behind this synthesis "art". In order to progress significantly, it is important to discover the mechanisms that allow predicting and controlling the self-assembly process. New creative synthesis routes could be developed that employ patterned spatio-temporal dynamics of templating chemicals and/or building units. Particularly inspiring in this regard are the variety of complex, yet structured pigment patterns that develop on the shells of mollusks, and that could be explained by nonlinear physico-chemical mechanisms as discovered by the mathematical biologist Meinhardt.

Search For Optimal Patterns For Transport And Reaction Problems

In the search for optimal patterns for transport and reaction problems, analytical and computational methods are invaluable.

A first methodology used in my group consists of hierarchical simulation techniques, which combine macroscopic continuum calculations with statistical mechanics at the microscopic scale. An example is schematically shown in Figure 8. I stress the essence of the methodology, not the details. The yield or selectivity of a real industrial catalytic process is affected by, in this example, the nm-scale roughness of catalyst pores, a scale more than a billion times smaller, but clearly significant. This effect can be studied by a combination of simulations at all relevant scales, enabling an optimization of the yield or selectivity. In order to do so, transport through the catalyst pores, often a limiting step in chemical reaction engineering, needs to be studied. Dr. Kourosh Malek studies this problem in my group using dynamic Monte-Carlo simulations, in which the trajectory of diffusing and reacting molecules is followed (Figure 9). The next step is to combine this with molecular dynamics.

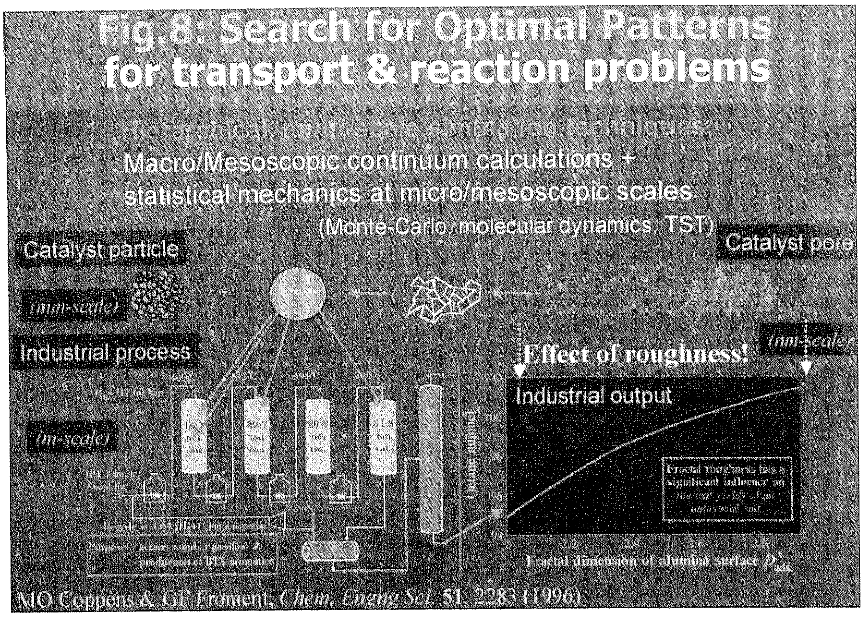


Fig.8: Search for Optimal Patterns
for transport & reaction problems
1. Hierarchical, multi-scale simulation techniques:
Macro/Mesoscopic continuum calculations +
statistical mechanics at micro/mesoscopic scales
(Monte-Carlo, molecular dynamics, TST)
Catalyst particle
(mm-scale)
Catalyst pore
(nm-scale)
Industrial process
(m-scale)
Effect of roughness!
Industrial output
Octane number
Fractal dimension of alumina surface
MO Coppens & GF Froment, Chem. Engng Sci. 51, 2283 (1996)

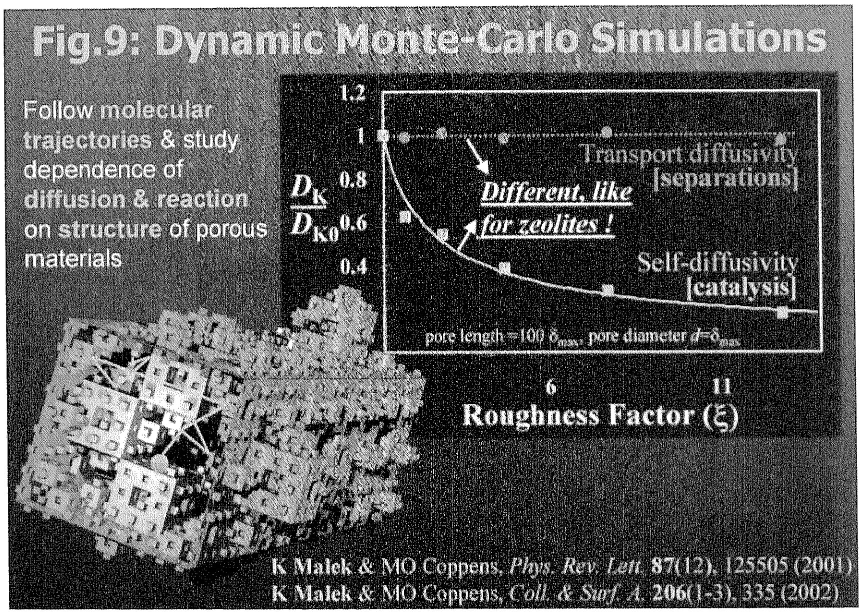
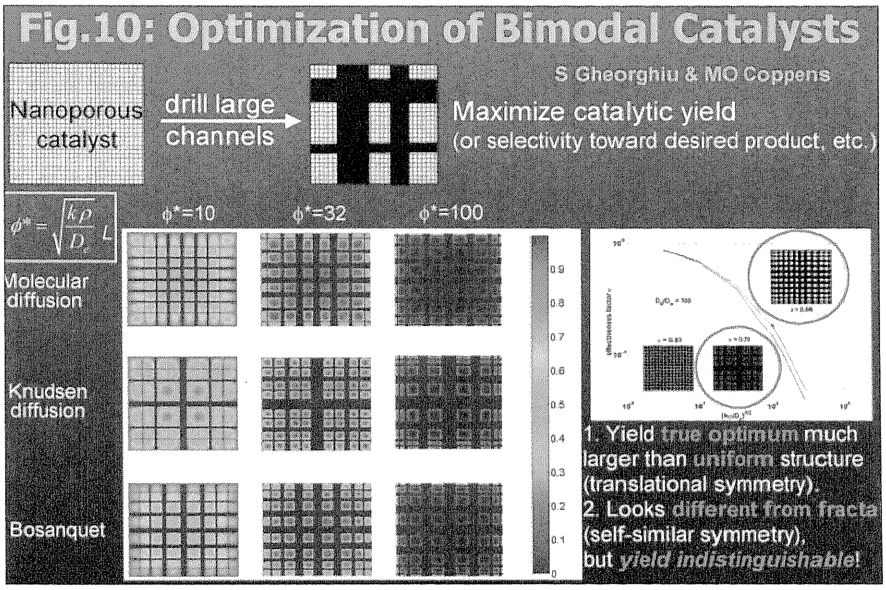


Fig.9: Dynamic Monte-Carlo Simulations
Follow molecular trajectories & study dependence of diffusion & reaction on structure of porous materials
1.2
1
0.8
0.6
0.4
D_K / D_{K0}
Transport diffusivity [separations]
Different, like for zeolites !
Self-diffusivity [catalysis]
pore length = 100 δ_{max}, pore diameter $d=\delta_{max}$
6
11
Roughness Factor (ξ)
K Malek & MO Coppens, Phys. Rev. Lett. 87(12), 125505 (2001)
K Malek & MO Coppens, Coll. & Surf. A. 206(1-3), 335 (2002)

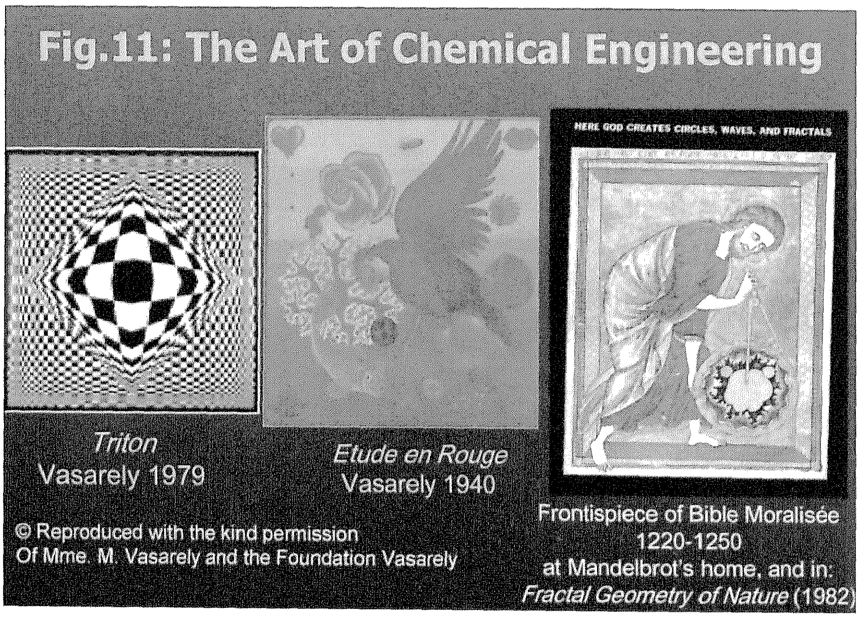
A next significant step in the optimization, to be used in combination with the synthetic methods discussed earlier, is the following (Figure 10). Large pore channels are drilled within a nanoporous catalyst. Molecules need to diffuse from the outside to the inside, where they react, and the idea is that the large "highways" will speed up the process in the "nano-alleys". The number, size and positions of highways can be changed at will. Required is a maximum yield, or, eventually, a maximum selectivity toward a desired product, or another optimum, depending on the problem at hand. My post-doc, Dr. Stefan Gheorghiu, found the following results for the maximum yield. Red corresponds to a high effective usage, blue to a low effective usage. Without discussing the details, it is clear that the optimal architecture depends on the process details. Encircled are the optimal structure for a particular relevant situation, on the right, and an optimized fractal structure. What we found is that (1) the true optimum has a significantly higher yield than a uniform classical structure with translational symmetry, pleading for a deviation from uniformity, while (2) the optimum may look quite different from a fractal structure but the yields are virtually indistinguishable, pleading for the use of a more easily synthesized, modular fractal structure.

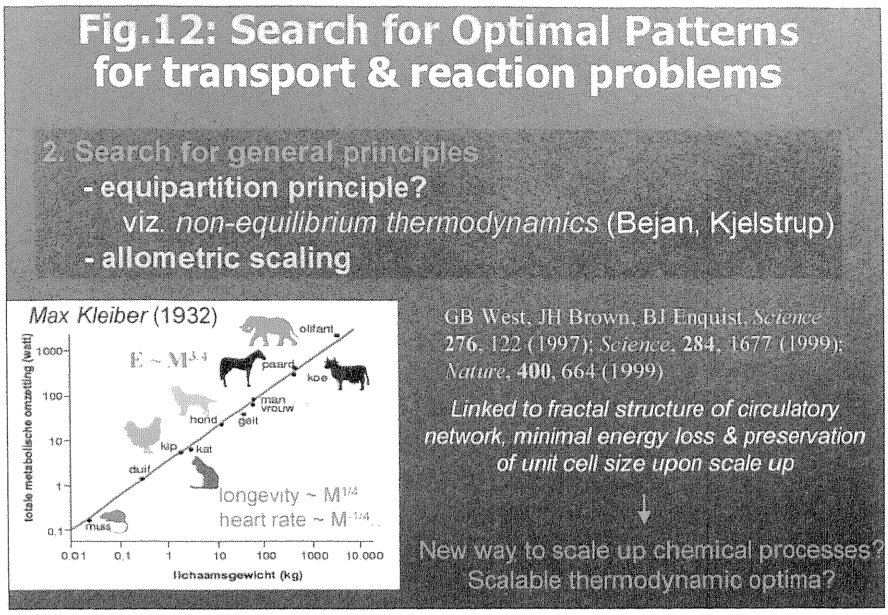


These patterns are esthetically very pleasing and immediately reminded me of something in Art (Figure 11). One of my old-time favorites, Victor Vasarely, the exponent of Optical Art or Op Art, painted many geometric patterns such as this *Triton* in 1979. I also found the following, older painting, *Etude en Rouge*, from 1940. Clearly more classical, but isn't this a fractal coral in the back? That fractals are part of Nature cannot be more beautifully displayed. And also this fractal looks very familiar, as a similar one appears on a frontispiece of a *Bible Moralisée* from the 13th century, to be found near the dinner table at Mandelbrot's home, and displayed in the *Fractal Geometry of Nature* as: *Here God creates Circles, Waves and Fractals.*

Let us now return to our search for optimal patterns. A second route I am only starting to explore is one for more general principles (Figure 12). This is potentially much more powerful than purely computational work: are there natural fundamentals behind the optimal patterns?

A first possibility is the existence of some sort of "equipartition principle," a concept explored by Professor Bejan at Duke and Professor Kjelstrup at Trondheim in the framework of non-equilibrium thermodynamics.





The equipartition of space, *e.g.*, naturally leads to optimum heat transfer, and predicts a fractal tree-shaped cooling fin to be optimal. Is there something similar for chemical reaction processes?

A second possibility is the existence of some sort of "allometric scaling." The zoologist Max Kleiber discovered in 1932 a remarkably universal scaling law: on average, the energy dissipation by an organism is proportional to the 3/4 power of its body mass. A classical description would predict a 2/3 power, because mass is proportional to volume, volume to length to the 3rd (m^3), and dissipating area classically to length squared (m^2). There are related scaling laws, such as a constant average number of heartbeats. Very recently, a group of researchers from the Santa Fe Institute showed that this law could be derived based on the fractal structure of the circulatory network in each of these organisms, in conjunction with a requirement of minimal energy loss and a preservation of the smallest capillaries upon scale-up. Indeed the cells or smallest units have the same size in, say, a mouse, a man or a dog. The physico-chemical function is related to this microscopic size, and only the macroscopic size, *i.e.*, the size of the organ or organism as a whole, differs. A space-filling fractal network of blood vessels links micro- and macro-scale, and it is this fractal nature that leads to the 3/4 law.

My suggestion now is to use this concept to find a new way to scale up chemical processes, and perhaps find scalable thermodynamic optima, in a similar way as in nature. This would provide a fundamental way to find the optimum hierarchical network linking the micro- with the macro-scale.

Interestingly, however, and this could be linked to our findings for porous materials, the structures of the branching tree itself and of the network of nerves inside the leaves on the tree are different (Figure 13). Similarly, the structure of the smallest hierarchies of airways in a lung differs from the large-scale self-similar hierarchy. It is proposed that the difference in dominating physico-chemical phenomena at different scales (*e.g.*, diffusion at small scales versus flow at large scales) leads to this discrepancy, and that the structure is therefore a more complex, composite one. We could once again learn from nature as a chemical engineer!

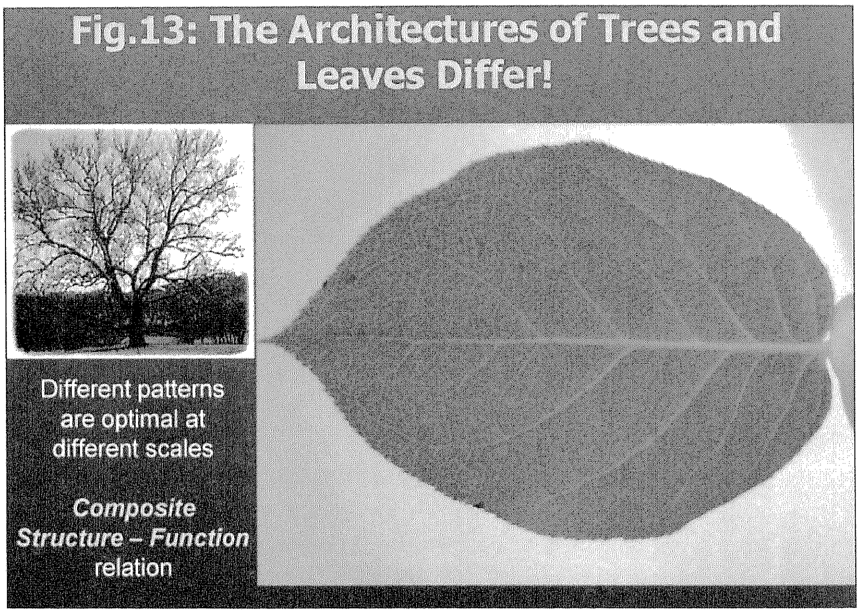


Fig.13: The Architectures of Trees and Leaves Differ!

Nature-Inspired Chemical Reactors

I will now move away from porous materials to my second topic: the nature-inspired rational design of chemical reactors and processes.

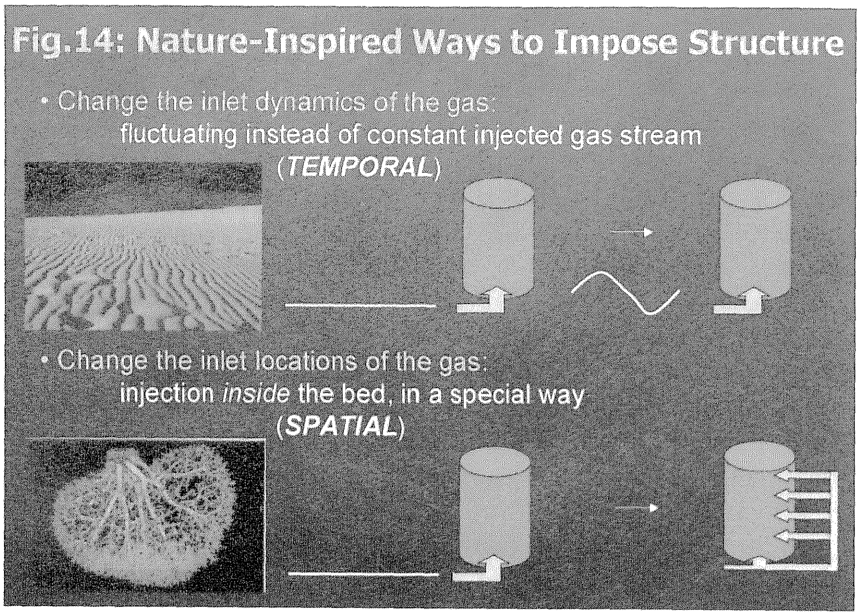
Earlier, I already showed two nice examples of structured reactors and distillation packings (Figure 3). The idea behind these structures, as investigated by my colleagues at Delft, is to replace random packings by controlled structured packings that could lead to desirable properties, such as a lower pressure drop, higher efficiency, and easier scale-up.

These are so-called fixed beds or static solid structures through which liquids and/or gases flow, which may, *e.g.*, react inside small solid catalyst particles that are coated on these structures. Many other multiphase reactors use only fluids or fluidized solid particles, and are intrinsically dynamic in nature. Would it be possible to also impose the aforementioned principles from the first example to structure also these reactors?

An important industrial example is a gas-fluidized bed of solids. In this reactor, a bed of reacting or catalytic solid particles rests on a distributor plate, through which a gas is injected at the bottom. When this upward flow is sufficiently high, the solid particles are fluidized and turned into an emulsion, the hydrodynamics of which is very complex. Typically, excess gas leads to the formation of bubbles. These bubbles rise, grow and coalesce. Professor van den Bleek and Professor Schouten demonstrated the chaotic nature of the hydrodynamics. Scaling a process up from a small to a large scale is complex, because the phenomena are scale dependent. Therefore, modeling, scale-up and control remain a challenge, and, despite significant advances in computational fluid dynamics, this field is still very empirical.

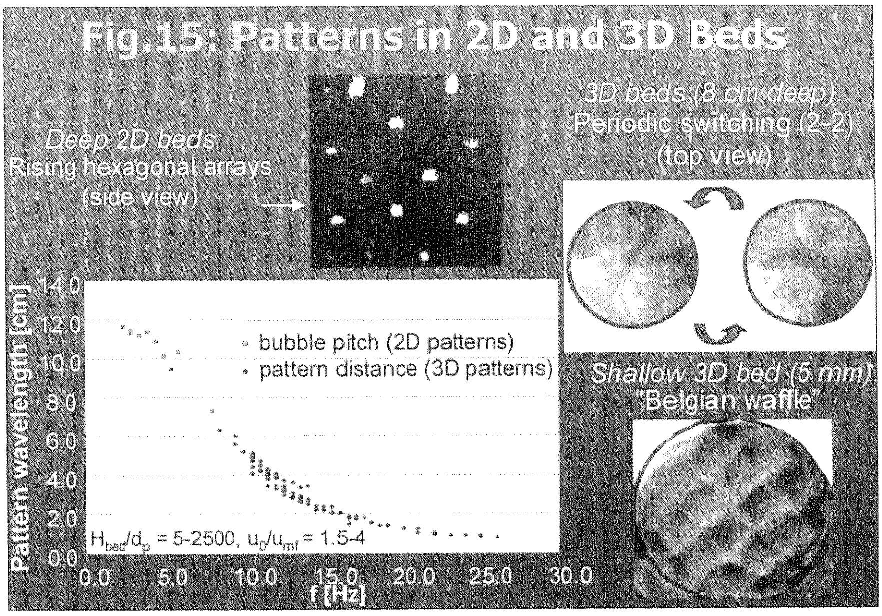
This calls for ways to *rationally structure* fluidized beds, to control the bubble sizes, make the bed more uniform and facilitate scale-up. I propose two fundamentally different ways to impose this structure – one temporal, the other spatial (Figure 14).

The first one is temporal, and is reminiscent of the translational symmetry of patterns on a sandy beach. The dynamic action of wind and water leads to these regular patterns. Also in a fluidized bed, we could inject the gas in an oscillating, fluctuating manner, instead of constant.



Videos [shown during the presentation] show what happens, first in a two-dimensional bed of sand particles. The first video shows the normal operation with constant airflow from the bottom. A two-dimensional bed is thin enough so that a light shining behind it reveals the bubbles; it is not possible to look through a fully three-dimensional bed. The second video shows what happens when the flow oscillates, at 2 Hz in this case. A clear order appears. This beautiful dynamic hexagonally ordered pattern is also seen in larger beds; moreover, the wavelength is independent of bed width, and exists up to a height approximately equal to the bed width, after which it becomes chaotic again. These properties point to a clearly nonlinear phenomenon, and not a simple resonance.

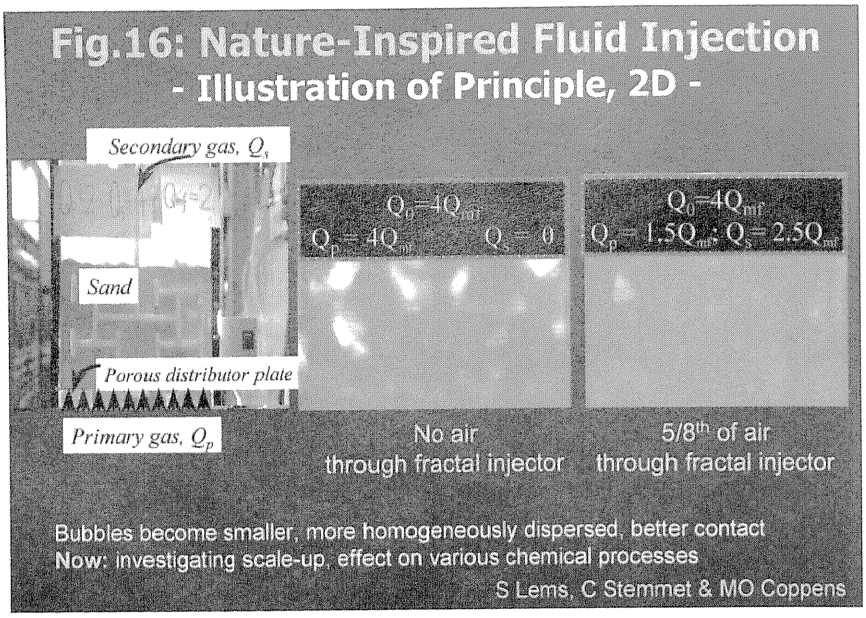
In 3D, looking from the top, since it is not possible to look through the walls, periodic switching and "Belgian waffle"-like patterns are observed. This pattern formation seems to be specific for multiphase flow, as it differs from other patterns in liquids or in granular media, where waves are damped after a few particle heights. Further experimentation and modeling should shed more light on this (Figure 15).



A second way to impose structure is once again inspired by the structure of a lung or kidney (Figure 14, lower part). Instead of only injecting gas from the bottom, a fraction of the gas is injected as secondary gas at different heights in the bed, but through a fractal tree-shaped injector.

The incentive behind this is that kidneys, lungs or trees, with their internal fractal self-similar symmetry, can be viewed as transfer devices (filters or oxygen-carbon dioxide membranes) or as reactors (photosynthesis). Since they have no characteristic scale except for the constant size of the smallest building units (capillary, twig or cell), these reactors and transfer devices of nature are trivially scalable: a small one, or a growing one, is similar to a larger one. They also have the earlier mentioned advantage of uniform and fast distribution of materials.

Let us again look at the fluidized bed shown earlier, now with the perhaps simplest (pre-)fractal tree-shaped injector suspended in it (Figure 16). Part of the gas is injected as primary gas from the bottom, while another part of the gas can be injected as secondary gas through the injector tube. The distance from the inlet to each of the 16 open outlets is the same, so that gas flows out of each outlet with almost the same velocity.



The result is clearly seen in Figure 16, on the left with only primary air, on the right with part of the air through the injector. When using the injector, for the same total amount of gas, the bubbles are clearly smaller, more homogeneously dispersed, with better contact between gas and solids. The next step is to investigate scale-up, and the effect on various chemical processes. It is particularly pleasant that Sander Lems, an alumnus of Professor de Swaan Arons (who is giving his valedictory address today), is now doing his Ph.D. with me on this topic. Figure 17 shows a scaled-up version, for a bed that is twice as wide and twice as high: consistent with scale-up in nature, the distance between the capillaries and that between the outlets is kept constant, only the number of generations in the hierarchy is changed.

In three dimensions, similar principles are applied. Shown in Figure 18 is an injector with fractal dimension 2.6, both for a large and a small vessel; the size of the outlets is the same. The fact that its dimension is fractional and lower than 3, means that it will not occupy a too large fraction of space, yet this design does allow gas to access the entire reactor cross-section.

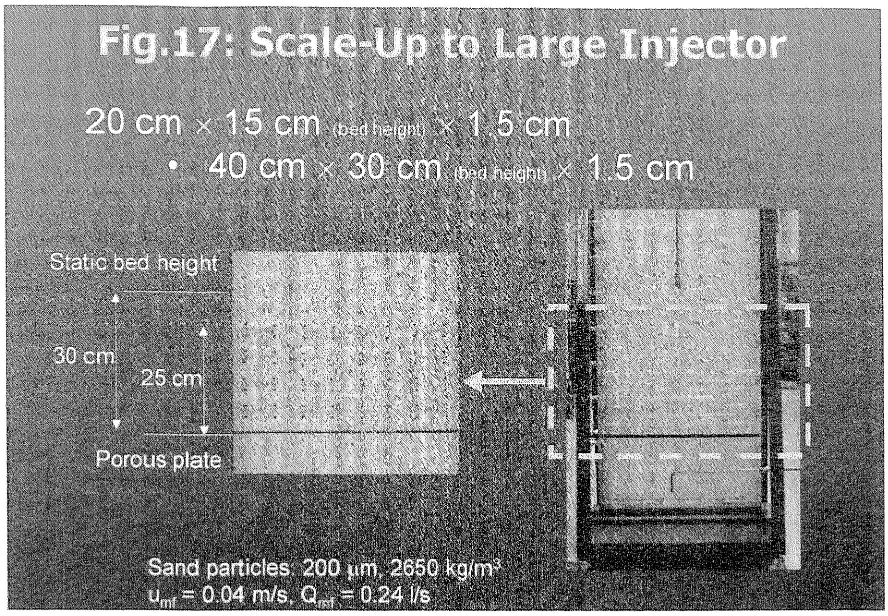


Fig.17: Scale-Up to Large Injector
20 cm × 15 cm (bed height) × 1.5 cm
• 40 cm × 30 cm (bed height) × 1.5 cm
Static bed height
30 cm
25 cm
Porous plate
Sand particles: 200 μm, 2650 kg/m3
umf = 0.04 m/s, Qmf = 0.24 l/s

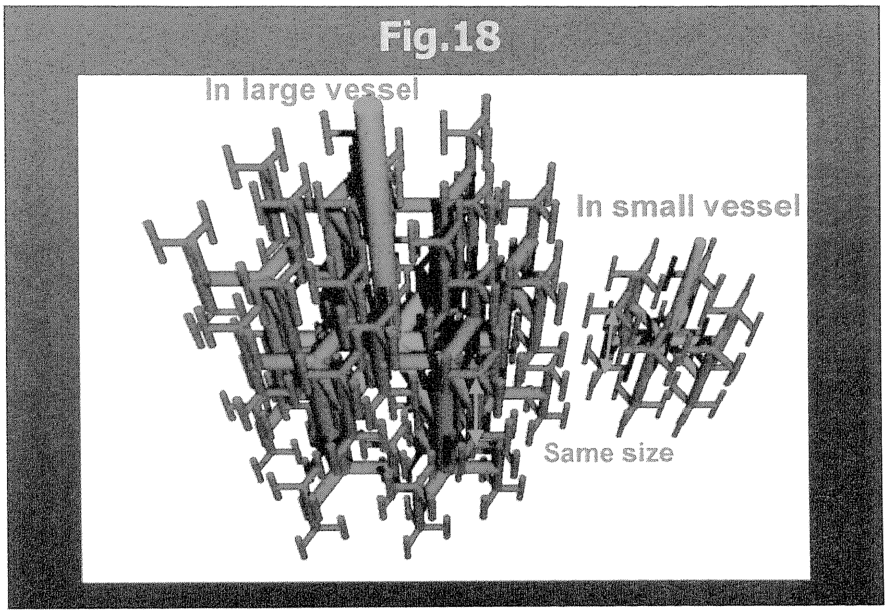


Fig.18
In large vessel
In small vessel
Same size

In nature, understandably, self-similar hierarchical structures that serve different functions have a different structure, *e.g.*, lungs and kidneys from different animals have a different fractal dimension. It is to be expected that the optimal hierarchical structure will also differ for different technological processes.

Search for Optimally Structured Multiphase Processes using NICE
The concept of the fractal injector is quite general, as it mimics various similar devices in nature. There are therefore many potential applications for gas/liquid, gas/solid, liquid/liquid, or slurry reactors, in the bulk or the fine chemical industry, for the polymer industry, in biotechnology (*e.g.*, fermentors), water purification (*e.g.*, aerators), absorption and mixing processes (viscous and non-viscous).

Advantages are the robust, scalable, flexible and natural modular design, the absence of moving parts, and the local control *via* gentle micro-mixing. A U.S. patent was issued for this device and its use.

An important challenge now is how to optimize fluid distribution in time and in space. Like in the design of porous materials, but even more relevant for scale-up, unifying principles such as allometric scaling will be searched for, and it would be useful to develop and apply fundamental thermodynamic principles similar to the equipartition rule. Above all, progress should be aided by a closer look at nature's reactors.

Conclusion
To conclude, recognizing the architecture of nature, learning how nature builds patterns and hierarchical structures, can lead to rational designs that improve the efficiency of chemical products and processes in a sustainable way.

Teaching

Having discussed research objectives, allow me to switch gears, and spend a few words on *teaching*. This is an essential element of my profession, one I care deeply for, and therefore devote a lot of time to. When preparing my classes, I often think of some of my own outstanding teachers, like Professor Kuliasko from Ghent, or my senior high school chemistry and mathematics teachers, Mr. Van Paemel and Mr. Goethals, respectively, and I cherish the advice I received from Professor Prausnitz at Berkeley, an excellent teacher himself. Genuine interest, being motivating and straightforward, aiming for accuracy yet also simplicity in passing on a subject, and stimulating creative thinking, are elements that I consistently try to implement in my own classes, now that I am standing in front of a class; no memorization but understanding and application; constant revision of classes; interaction with students individually and in groups. There is great pleasure in teaching, encouraging and communicating with young students, and to pass on a passion. One appreciates a subject best when teaching it.

Thank You

One important reason for me to give this oration is to express my gratitude. There are many people that I am immensely grateful to for their support throughout the years, people who have had a significant influence on my education, past and current development, either directly or indirectly. I see science as an art, and, as my friend John Prausnitz, engineering as a humanity. Therefore, I benefited tremendously from the constant encouragement and the interactions with my artistic open-minded parents, as well as from discussions with my beloved wife Cynthia. If I have yet another crazy idea, Cynthia is the first person I talk to, for her frank opinion is typically accurate, genuine and down-to-earth, and it is invaluable to get the insight of somebody who is creative and open-minded, but not in the field. Cynthia, I know you did not want to be mentioned in my speech, because you don't feel I owe you anything academically, but I am stubborn and do so anyway, because you're very precious to me in every respect.

At this moment, I would like to gratefully acknowledge a number of people who are instrumental in the academic work I discussed today. The oration protocol discourages thanking a long list of personal friends separately at this time – and a long list there is –, however I insist on the pleasure to thank you personally at the reception following the ceremony, or at the dinner party tonight.

I express my appreciation to the Board of the University and to the appointment committee for their confidence in me, by appointing me to a full professorship. Special thanks to the department chairman, Professor Gijs Ooms and Professor Jacob Moulijn, for their constant support, as well as to Dean Karel Luyben, people who made it possible for me to start this unconventional research program by providing support in the early stages when I really needed it.

Furthermore, my research could not be carried out without my invaluable collaborators and research team – present and past post-docs, Ph.D. students, Master's students, and technicians. A special word of thanks to Mrs. Els Arkesteijn, our management assistant, for her infinite patience, sense of detail, good humor, and help with the organization of a sometimes proverbially absent minded professor.

King Leopold II of Belgium once said *La musique est un bruit qui coute cher*; the same can be said of scientific research – an "expensive hobby": its use, like beauty, is in the eye of the beholder, and I am indebted to those people that see its use. I am grateful to the Dutch Foundation of Scientific Research (NWO) for giving me considerable support, valued at more than one million Euros, through Open Competition grants, a Young Chemist award and, most recently, a PIONIER award. Full sponsorship from Akzo Nobel Catalysts for a Ph.D. project of a more fundamental nature is valued highly in these times where the application time horizon of many companies has shortened. I have not forgotten my *alma mater* Ghent University, the National Foundation for Scientific Research FWO-Vlaanderen, and the Belgian American Educational Foundation, who funded my early research, both in Belgium and in the United States.

My time as a PhD student with Professor Froment has been instrumental in my scientific development, and it is then that I truly learnt the importance of application-oriented research. Professionally and personally, I also benefited greatly from the mentorship of Professors Mandelbrot, Bell, and Prausnitz, to an extent I will elaborate on later tonight.

Professor van den Bleek, dear Cor, the last words of gratitude in this oration are addressed to you, as you brought me here to Delft. I did not know exactly which adventure I would embark on when I accepted your offer. Despite opportunities in the United States, a tour of the lab and conversations with you, Jacob Moulijn, your colleagues, and several of your students, convinced me to come to Holland. I vividly remember my first visit. I was impressed, and stayed impressed. It is both a pleasure and a privilege to be on the faculty of this excellent department. But it is a one-in-a-million chance to work with you. Your greatest pleasure is when people working with you succeed, and you always put your collaborators or section members in the spotlight, independently of whether you participate in a project or not, giving them credit whenever the opportunity arises. You encouraged my unconventional ideas, and you applauded that I started various new projects and developed my own vision. But it was the environment that you created that made the implementation possible. Your visionary work on chaos in fluidized beds matches well with my ideas on structuring chaos. I learnt a great deal from you, and there is no doubt that your support and motivation made it possible for me to have such a fast start and thrive here in Delft.

Let me finish by stressing that I see today especially as a moment to look forward. Self-indulgence or pride would be out of place, as my oration marks something of a beginning, accepting to take on extra responsibilities: The more titles one accepts to carry, the heavier the responsibilities. I am full of optimism, ready and glad to take them.

I have avoided giving a too detailed overview of all the research topics I am working on now, or the ones I will work on in a few years. The latter, I am unable to, as I do not know where the adventure will lead my group to, or which new challenges lay ahead. Knowing this would defeat the purpose, and much of the joy. However, I have tried to convey the philosophy behind my work, and my reasons to be here. The first poem my wife ever sent me was the Ode by Arthur O'Shaugnessy: *We are the music-makers, and we are the dreamers of dreams.* It is a lot of fun to dare to dream and wander, and to take a harmonious, nature-inspired road together. Let's move on!