

Network of Excellence



# **Editorial**

Welcome to the first issue of the SoftComp Newsletter.

Early this year, in a first step, we had implemented, the SoftComp web site as a major information channel for the dissemination of interdisciplinary research efforts, with support from the European Union's Sixth RTD Framework Programme. In a second step, this SoftComp Newsletter has been set up as an equally important information channel intended to reach, in addition to the SoftComp participants, a broader, more general audience.

To serve this purpose, the SoftComp NoE Newsletter is structured to inform its readers by overview articles on work performed, and scientific articles on specific SoftComp research topics. One popular scientific article will be dedicated to attracting a more general audience to the exciting issues of the SoftComp research efforts of finding tomorrow's solutions to today's problems.

Current SoftComp Network information will be communicated through short columns like Personalia, Coming up and Vacancies.

This Newsletter is intended to be issued twice a year.

For those of you not yet familiar with the SoftComp Project, but who are becoming more interested while reading the Newsletter, please visit our Web Portal: www.eu-softcomp.net

Hugo Bohn & Dieter Richter

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# The First 18 Months of SoftComp

Dieter Richter · SoftComp Network Coordinator Forschungszentrum Jülich · Germany

The Network of Excellence (NoE) Soft Matter Composites – An Approach to Nanoscale Functional Material (SoftComp) was founded with the aim of establishing a knowledge base for the intelligent design of functional and nanoscale soft matter composites. This is achieved in overcoming the presently two fold fragmentation of this important field for the development of new materials at the interface of non-living and living matter, where the delicate principles of self-organisation are ubiquitous. At between the various participants who provide expertise in a broad range of synthesis technologies for new materials, experimental facilities and methods as well as computational algorithms and analytical theory. Consequently, the integration concept of SoftComp is based on the development and strengthening the cooperation between the partners by the joint utilisation of devices, equipment and know-how in fields that are of the utmost interest for all partners. In this spirit, in the first year of

Figure 1



present soft matter is a multidisciplinary research topic along the disciplinary lines of theoretical and experimental physics, computational science. chemistry and applied biology. Soft matter materials range from polymers to colloids, from liquid crystals to membranes, from amphiphilic molecules to biopolymers (Figure 1). SoftComp aspires to create an interdisciplinary research platform dealing with this wide variety of soft matter materials. The Network unites a sizeable fraction of the very best European research groups and links these academic groups with some of Europe's most important industries dealing with such materials.

Synergies and added value are achieved by sharing resources and knowledge

SoftComp three research platforms were established providing the partners with the combined knowledge and techniques available within the network.

1. The Synthesis Platforms combine major laboratories for the synthesis of soft materials and their characterisation within the Network. Through this platform the experimental groups are provided with well characterised materials tailored for the specific needs of the various scientific workpackages. Even in the first 18 months the new opportunities created by the Synthesis Platform were already widely used and have initiated a common basis for materials throughout the Network.



# The First 18 Months of SoftComp

2. The *Experimental Platform* pools the speciality non-commercial equipment for soft matter research that is available within the NoE. In thus emphasis the Centres of Excellence in certain laboratories which are leaders in their specialities. The SoftComp partners are increasingly aware of these novel opportunities for broadening their research and using them correspondingly.

**3.** In the *Theory and Simulation Platform* a wide range of theoretical tools for treating soft matter problems are available and cross boarder activities between different groups are emerging.

In the first SoftComp year, a formal agreement between the SoftComp partners on ensuring sustainable scientific cooperations through the common use of the network infrastructure was concluded. Being aware of the importance of the platforms, the Network Governing Board (NGB) implemented a focused funding strategy to enhance the capabilities of these platforms and to strengthen their role in the Network. A first round of proposals was accepted and nine grants were given to partners in the Experimental and Synthesis Platforms.

In order to achieve the increasing integration and participation of industry, an International Advisory Board (IAB) with high level-industrial participation supervises the planning of SoftComp so that industrial aspects play an important role. As an example, we would like to emphasise that in the first year one company launched a new project relating to the phase and permeability behaviour of the stratum corneum (outer skin) which was taken up with great interest by the academic partners and led to a European proposal for further funding.

Scientifically SoftComp spans a broad range with more than 120 researchers actively involved. In this short overview it is therefore impossible to even attempt to present all important results.

### Figure 2



Entangling and merging worm-like micelles under shear.

Instead a few highlights are presented as an exemple.

Figure 2 displays a microscopic view of a new mechanism of fusion of two worm-like micelles under shear flow. The results were obtained by computer simulation and shed some light on the mechanism behind the strong shear thinning behaviour of such systems which are important candidate materials for use in tertiary oil recovery. The upper picture shows two entangled worm-like micelles preventing each other from flowing. Under shear stress the micelles gradually merge and are transformed into an H-shaped structure. In this state, the entanglement effect is



removed and the system flows easily (Twente/Schlumberger).

Figure 3 shows the phase separation in giant micelles containing a mixture of two lipids and cholesterol (sphingomeylin/cholesterol/DOPC). Such mixtures are biomimetic models for multicomponent biological membranes. The different phases are made visible by fluorescence microscopy using fluorescent markers. Taken at different compositions the results allow the determination of the phase diagram (Marie Curie/Bordeaux).

# Figure 4

Snapshot of the confinement effect within a miscible polymer blend of components with very different glass transition temperatures  $T_g$  (red: low  $T_g$ ; blue: high  $T_g$ ).

Figure 4 presents a snapshot of a polymer blend where the dynamics of the component with a low glass transition temperature ( $T_g$ ) (red) are dynamically confined by the frozen matrix of a high  $T_g$  component. This schematic picture visualises an experimental result where for the first time such confinement effects were observed directly by neutron scattering and also by computer simulation. These results may be important for designing new plasticises which would serve their main purpose without weakening the material (San Sebastian/Jülich).



# From Mud to Worms... Soft Composite Materials for Hydrocarbon Recovery

Edo S Boek · Schlumberger Cambridge Research · UK and Geoffrey C Maitland · Imperial College London · UK

Soft composite materials play a fundamental role in many aspects of oil and gas production processes. This is manifested not only by the mobilisation of multicomponent viscous and semi-solid hydrocarbon mixtures through displacement processes in porous rock media having throat sizes in the colloidal size range, but also by the use of complex colloidal mixtures in all phases of the recovery process. The industry, and Schlumberger in particular, has been actively involved in developing new, smarter soft composite materials, both to increase the efficiency of constructing wells of lengths in excess of 12 km and to improve reservoir hydrocarbon recovery factors from the 30-40% typical of today to targets of 60-70 % over the next decade.

# Drilling oil and gas wells

The first stage of recovering the oil and gas from an underground reservoir is the drilling of wells into hydrocarbonbearing rock (see Figure 1). This is performed by rotary drilling using a drilling fluid, or *mud*, that must perform many functions, including having the right rheology, forming a lowpermeability protective filtercake on the freshly created wellbore surface and having suitable physicochemical characteristics to avoid chemomechanical failure of weak colloidal clay rocks, swelling shales. One major class of these muds is based on suspensions of smectite clay colloids (montmorillonite). Others use biopolymers or water-in-oil invert emulsions to give the highly shear-thinning viscoplastic rheology



Typical rig and well arrangements for onshore and offshore oil and gas production Wells are drilled in increasingly deep water, they have increasingly complex geometries (extended horizontal sections, multilateral configurations) to access more of the oil and gas and data are transmitted by satellite to and from the rigs to enable remote control and optimisation of production. [Reproduced by kind permission of Schlumberger from Oilfield Review. Winter 2002, p.15]

needed to suspend drilled rock cuttings in a soft gel when static, whilst enabling the fluid to be pumped with low pressure drops to the bottom of the well and back.

Recently, composite colloidal fluids have been developed using hexagonal cationic nanoplatelets of mixed layer AI-Mg hydroxides in combination with larger anionic smectite clays to give thixotropic fluids which gel extremely rapidly and reversibly at rest but have viscosities of only a few centipoise at the shear rates experienced in the well (~50-800 s<sup>-1</sup>). The filtercakes formed on the porous rock walls from drilling fluids (see **Figure 2**) are dense soft composites made from mixtures of colloidal particles, polymers or emulsions, whose mechanical, permeability and adhesive characteristics are critical to sealing the well during the drilling process.



SEM image of a filtercake formed by pressure filtration of a water-based drilling fluid on a porous rock (Photo: John Cook, Schlumberger). The compact is a soft composite of montmorillonite clay platelets, which are stacked into flexible, parallel arrays, wrapped around larger barytes (BaSO<sub>4</sub>) particles added to the fluid to increase its density. The cake has an extremely low permeability (~1 nanodarcy, of a typical sandstone rock of 50 millidarcy). The cake yield stress is of the order  $10^5$  Pa and increases with clay volume fraction  $\phi$  in a power-law manner with an exponent ~3 which extends over concentrations from dilute muds ( $\phi \sim 0.05$ ) all the way to cake compacts ( $\phi > 0.5$ ) (John Sherwood, Gerry Meeten, Schlumberger Cambridge Research).

# The problem of swelling shale rocks

The most troublesome rocks to drill through are shales, a natural soft composite composed of clays, silt and other minerals filled with saline pore fluid. Understanding the physical chemistry of the osmotically-driven swelling of weak shales, containing smectite clays, by water-based muds (which can cause major, potentially catastrophic stability problems in newly-drilled wells) has been a major focus of study. Molecular simulation, FTIR surface spectroscopy and



thermodynamic modelling have all played a role in the molecular design of short-chain polyalkoxide copolymers which can inhibit this swelling process by adsorbing on the rock clay surfaces, displacing hydration water and forming adsorbed bilayers stabilised by hydrophobic interactions – polymer clay composites.

# Stimulating production from the reservoir

Once a well has been constructed, its production can be limited by low natural reservoir permeability or by permeability reductions induced by colloidal particulate damage to the rock pore space either during drilling or production. The creation of hydraulic fractures is one major stimulation process commonly used to overcome these limitations. This requires the use of viscoelastic gelling fluids to transport sand particles uniformly through the long (~300 m) fractures as they are formed, to keep them open after the hydraulic pressure is removed. Traditionally natural polymers, such as guar cross-linked by borate or transition metal complexes, have been used. However, even after degradation by oxidising breakers, these materials still block the pores in the sand-filled fractures, which therefore do not enhance production as much as anticipated.

Recently new surfactant-based aqueous fluids have been developed which form strong viscoelastic gels, through entangled worm-like micelles during fracture creation, but break down to much smaller spherical micelles with low viscosity once hydrocarbon starts to be produced through the fractures (see Figure 3). This enables an essentially



oil production can be limited due to radial flow into the well. The hydraulic creation of two opposing vertical fractures (bottom) extending up to 300m from the well gives large productivity increases due to linear flow into the highly conductive fracture.
(b) Cryo-TEM image of the worm-like micelle structure obtained for a 4.5 %

 $C_{22}$  quaternary ammonium cationic surfactant in 2 % potassium chloride, typical of the viscoelastic surfactant systems being used as fracturing fluids. This structure breaks down to spherical micelles or microemulsions as hydrocarbon is produced through the proponant pack complete clean-up of gel from the fractures, leaving a porous sandpack in place to give maximum production enhancement. Composite materials combining surfactants, polymers and colloidal particles can be used to tune the balance of properties required for particular conditions.

# The role of computer simulation

A key enabling science in the design of these complex materials has been computer simulation. The aim is to bring a systematic design methodology, all the way from molecular structure to bulk material properties, to the materials formulation process, which is notoriously based on much empirical selection of components and composition within the vast parameter space available to the designer. Advances in mesoscopic simulations which bridge the length scales from molecular dynamics to macroscopic CFD







are now beginning to enable the effects of changes in molecular and colloidal characteristics on bulk rheology to be predicted, at least at the trend level (see Figure 4, which compares experimental and predicted behaviour for worm-like micelle fluids).

# Smarter responsive materials

The development of such smart, responsive soft materials, whose physical properties can be transformed *in situ* by natural process triggers, such as solvent change, temperature or shear, has been a major research focus in recent years. Other examples include thermoassociative polymers (which maintain essentially constant rheology over wide temperature ranges through hydrophobic associations of side chain branches which exhibit an LCST) and shear-induced gels for blocking large fractures in drilled rock (which form *in situ* by the shear-induced destabilisation of an emulsion – having polymer in one phase and cross-linker in the other – as the fluid passes at high shear rate through the nozzles of a drill bit).

# Soft composite materials are everywhere...

Research into soft composite materials at Schlumberger impacts many other areas. In cements, colloidal particles are used alongside the coarser cement grains to enhance the solids packing fraction to as high as 0.9, to enhance both slurry rheology and the mechanical permeability characteristics of the set material; a detailed knowledge of depletion phenomena is required to control such systems. The control of crystal growth processes by the blocking adsorption of molecules whose motif matches the crystalline arrangements of fast growing faces has been studied to design cement hydration retarders and inhibitors to prevent the formation of damaging inorganic scales such as barium sulphate. The characterisation, inhibition and control of colloidal asphaltenes and waxes in crude oil is key to limiting the permeability damage and emulsion formation caused by such materials during oil production.

The list could go on for some time; there is hardly any aspect of hydrocarbon recovery that avoids the attention of scientists working on soft materials. These examples illustrate the diversity of applications and the links of the underpinning science to that used in many other parts of the industrial formulation sector. Yoghurts and drilling fluids have a lot in common! However the challenges in the oil industry are to implement solutions which can operate reliably up to 250-300°C, 1000 bar pressure and be controlled remotely, many kilometres away from their point of injection. So oil field soft composites need to be tougher as well as smarter...but they're getting there.

# **About SoftComp**

**SoftComp** is a Network of Excellence – a tool developed under the 6th Framework Programme of the European Commission dealing with the integration of European research, with the intention to strengthen scientific and technological excellence. In particular, SoftComp aims to establish a knowledge base for an intelligent design of functional and nanoscale soft matter composites. It will do so by overcoming the present fragmentation of this important field for the development of new materials at the interface of non-living and living matter, where the delicate principles of self-organisation in polymeric, surfactant and colloidal matter are ubiquitous. This Network of Excellence will create an integrated team that is able to activate the European potential in soft matter composite materials and thus disseminate excellence through extensive training and knowledge transfer schemes.

**SoftComp partners**: www.eu-softcomp.net/about/part **SoftComp funding programme**: During the past 8 months more then 100 applications for grants were submitted. **Registration**: Presently 130 participants have registered. If you want to register contact: f.h.bohn@fz-juelich.de (5)

The following Conferences were sponsored by SoftComp in 2004/05.

Conference/Place	Date	
Conference: Jülich Soft Matter Days 2004 Kerkrade · The Netherlands	16-19 November 2004	
Conference: SoftComposites Forming Reversible Networks · Montpellier · France	18-19 November 2004	
X-ray Microscopy Lab Course for Soft Nanostructures and Colloids Berlin · Germany	09-12 February 2005	
School: Introduction to Soft Condensed Matter and Advanced Colloid Science Utrecht · The Netherlands	28 February - 11 March 2005	
Workshop on Critical Questions and SoftComp Strategies for Long Chain Branched Polymers Leeds - UK	22 March 2005	
<b>Workshop on Dynamics in Polymer Blends</b> San Sebastián · Spain	02-04 June 2005	
Workshop: Materials Simulation Days 2005 Mainz · Germany	08-10 June 2005	
6 <sup>th</sup> EPS Liquid Matter Conference Utrecht · The Netherlands	02-06 July 2005	
European School on Rheology: Rheological Measurements Leuven · Belgium	12-16 September 2005	
9th Laboratory Course on Neutron Scattering Jülich · Germany	12-23 September 2005	
Workshop on Soft Interfaces with Hydrodynamic Interactions Amsterdam · The Netherlands	29-30 September 2005	
Multiscale Modelling in Biological Systems Conference · Salt Lake City · Utah · USA	05-07 October 2005	
Conference: Jülich Soft Matter Days 2005 Bonn · Germany	01-04 November 2005	



# Long-Chain Branched Polymer Dynamics

Tom McLeish · Department of Physics & Astronomy · University of Leeds · UK

A subset of the SoftComp NoE partners has begun a collaborative project on unravelling the subtle science of long-chain branched (LCB) entangled polymer melts. The first clue that the overall *topology* of polymer melts might alter their dynamics and rheology radically came with the discovery that the highly branched low-density polyethylene possessed special *strain hardening* properties in extensional flow that linear molecules did not. Work on monodisperse materials in the laboratories of the Polymer IRC at the University of Leeds, together with synthesis in Jülich, Durham and Sheffield, has confirmed and clarified these findings. It is an example of the way in which highly monodisperse or controlled-architecture polymers refine molecular thinking and theories of soft matter far better than simply working with industrial materials.



In Figure 1, linear rheological spectra from a linear and star-branched polyisoprene of identical *span* molecular weights are compared. The spectrum of the star polymer melt is much broader than the linear, and the terminal relaxation time is four orders of magnitude greater! The physical reason for this emerges naturally from the *tube model* of Doi and Edwards: star polymers cannot *reptate* in their tubes to relax their configurations like linear polymers but instead must exploit exponentially rare retractions of the arms, so that at long timescales the relaxing object looks like a slow central star-like core surrounded by rapidly moving and reconfiguring outer material (see Figure 2).

Even more striking are the rheological spectra of *H*-polymers or combs. These have been under intensive investigation by three of the SoftComp groups: Leeds, Jülich and FORTH. Their spectra, although of exactly monodisperse materials, show two regions of relaxation: a medium-frequency shoulder of relaxations that come from dynamics of the star-like arms of the polymers, and a sharp low-frequency peak that



corresponds to linear-like reptation of the cross-bar or *backbone* piece of the polymer (see Figure 3). The really remarkable aspect of entangled LCB melts rests, however, in their non-linear properties. This is the key to their industrial application in stabilising otherwise highly unstable extensional flows. The molecular roots of this *strainhardening* behaviour lie in the dynamics of the branchpoints themselves under high strain. At first, theory and experiment predict that they will act as cross-links of a rubber (*rubberlike* is sometimes used for *strain-hardening*),



but that beyond a critical strain the branch points are dragged into the tubes belonging to the branch nearer to the centre of the LCB molecule. Thus the material carefully codes for the strain hardening, and the critical strain at which the hardening should switch off - allowing the melt to flow! This phenomenon has stark consequences for molecular configuration as well as for rheological response, and has been the subject of a SoftComp collaboration between the Leeds and Jülich groups in using and modelling small angle neutron scattering (SANS) from strained melts.





Figure 4 shows a series of SANS 2d-intensity plots versus scattering vectors q from a melt of pure H-polymers, deuterated only in their cross-bar sections, made by J. Allgaier. They are strained by a factor 2 in the neutron beam (experiment by Wim Pyckhout-Hinzen). Note that, remarkably, as the relaxation proceeds in time (via careful heating and cooling of the material across its glass transition temperature in the neutron beam) across the upper series of plots, the anisotropy actually *increases* as a function of time, even though the stress is decreasing. These data are modelled very accurately (theoretical results in lower series of plots by Daniel Read) by assuming the retraction mechanism above, but this requires the additional re-arrangement of the relaxing arms into regions of lower elastic modulus. So SANS becomes a probe of physics invisible to rheology alone.



Comb materials behave in a similar way, and have now been synthesised in quantities large enough to measure extensional rheology and even complex flow fields, in a large UK-based collaboration *Microscale Polymer Processing*. Figure 5 shows both shear and extensional transient rheology of a pure polybutadiene comb synthesised by C. Fernyhough and measured by David Groves. The rheology is modelled by the *pom-pom* constitutive equation developed at Leeds. This then allows the complex flow of the monodisperse material



to be measured and compared to theory, when the physical rheology model is embedded in a viscoelastic flow-solver capable of handling viscoelastic models. Figure 6 shows a simulation compared to experiment (see Figure 5) of the comb melt in a constriction flow, visualised in the Cambridge laboratory of Malcolm Mackley, and simulated by Nat Inkson in Leeds. The down stream stress features emerging from the re-entrant corners are characteristic of LCB polymers, indicating that our understanding is now beginning to connect the molecular and macroscopic worlds of these materials.





# **Personalia**

Prof. Juan Colmenero, Universidad Euskal Herriko, San Sebastian, Spain, was awarded the Medal of the Spanish Royal Society of Physics.

Dr. Stefan Egelhaaf, University of Edinburgh, UK, accepted a professorship in the Department of Physics of Heinrich-Heine-University Düsseldorf, Germany.

The Royal Dutch Academy of Sciences announced the election of Prof. H.N.W. Lekkerkerker as Academy Professor.

Prof. Geoffrey Maitland, Scientific Advisor with Schlumberger, accepted a professorship at the Chemical Engineering Department of the Imperial College, London, UK.

Prof. Dieter Richter, Jülich, Germany, received and declined an appointment to the Governor's Chair of the University of Tennessee in Knoxville, connected with a position as Director of the Joint Institute for Neutron Science at SNS, ORNL, USA. He also was elected member of the Advisory Board of the Soft Matter Journals of the Royal Society of Chemistry, UK. Furthermore, he was nominated Chairman of the Scientific Council at ILL, Grenoble, France.

Dr. Didier Roux, CNRS Bordeaux, France, accepted a position as R&D Director of the Saint-Gobain Group, France.

Dr. Michaela Zamponi, Jülich, Germany, was awarded the Jülich Leibfried Prize for her presentation Polymers - Molecular Escape Artists, related to her PhD thesis work.

# Vacancies

### A full time tenure-track position...

...in the field of chemical product design is available at the Department of Chemical Engineering (CIT) of the Faculty of Engineering of K.U. Leuven, Belgium. More info: www.kuleuven.be/admin/rd/ niv3p/vzap6/ad-i02twt.htm Contact: jan.vermant@cit.kuleuven.be

### Three PhD positions...

...available at the Condensed Matter Theory Group in Mainz, Germany, in the area of simulation of colloidal systems in external fields or confinement. Contact: Dr. J. Horbach Email: horbach@uni-mainz.de

### **Open position...**

...in the Department Polymer Physics of BASF, Germany Profile: PhD in Physics / Chemistry / Physicochemistry Expertise in: colloid science, functional polymers, crystallisation, particle formation. Contact: Dr. Jens Rieger, BASF AG, Polymer Physics, GKP-G201, 67056 Ludwigshafen, Germany Email: jens.rieger@basf-ag.de

# PhD position...

....in the field of nanocomposites of polymers and nanoparticles, at Jülich, Germany. The planned thesis is embedded in the European NoE SoftComp and will represent a multiphysics approach by combining neutron and X-ray scattering methods with macroscopic dynamic mechanical techniques. The PhD position is available immediately.

More info: www.eu-softcomp.net/news Contact: W. Pyckhout-Hintzen, Research Centre Jülich, IFF (Neutron Scattering), D-52425 Jülich, Germany

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# Coming up...

Conference/Place	Date	
Annual SoftComp Meeting Venice · Italy • Joint NA-Meetings • EU Report Meeting	<b>02-05 May 06</b> 02-03 May 06 04-05 May 06	
IAB Meeting Paris · France	19-20 May 06	
EU Review & Assessment Meeting Düsseldorf · Germany	09 June 06	
37th Spring School 2006: Computational Condensed Matter Physics - Jülich - Germany	06-17 March 06	
10th Laboratory Course on Neutron Scattering Jülich · Germany	13-24 March 06	
Workshop on Mesoscopic Simulation Techniques for Soft Matter Systems Jülich · Germany	05-07 April 06	
2 <sup>nd</sup> International Workshop on Dynamics in Viscous Liquids Mainz · Germany	09-12 April 06	
Annual European Society of Rheology Conference Hersonissos · Crete	27-30 April 06	
The European School on Scattering Methods Applied to Soft Condensed Matter Bombannes · France	10-17 June 06	
3 <sup>rd</sup> International Workshop on Dynamics in Confinement Grenoble - France	23-26 June 06	
Ampere XIV NMR School-Soft Matter Poznań· Poland	22 June - 01 July 2006	
Summer School on Physics of Biological Objects Cargese · Corsica	14-25 Aug 06	
<b>Jülich Soft Matter Days</b> Bonn · Germany	15-17 Nov 06	
SoftComp SoftComp sponsored		

# **Credits/Disclaimer**

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