



INTERNATIONAL WORKSHOP ON

**Flow, Fracture,  
Interfaces  
in Soft Heterogeneous  
Materials**

**DECEMBER 9 & 10, 2013**

THE MICHELIN MATERIALS SCIENCE CHAIR  
AT ESPCI PARISTECH

# Programme

## December 9th

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**09:00**

*Jacques Prost (ESPCI ParisTech)*

Opening and introduction

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**09:20**

Keynote: *Maude Portigliatti, Fabien Gaboriaud (Michelin)*

“Design of reinforced soft matter in tire applications: a physical tricky compromise between rheology, fracture and tribology”

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### SESSION 1: Flow of Soft Materials

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**09:50**

*David Pine, New York University (USA)*

“Rheology and non-equilibrium phase behaviour of non-Brownian suspensions”

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**10:35**

Coffee break

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**11:00**

*Olivier Pouliquen, Polytech Marseille (France)*

“From dry to immersed granular flows”

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**11:45**

*Gareth H. McKinley, MIT (USA)*

“Quantitative Modeling of the elasto-visco-plastic flow of waxy crude oils and other soft thixotropic solids”

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**12:30**

Lunch break: Buffet and poster session

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**16:00**

*Peter Olmsted, University of Leeds (UK)*

“Fracture in entangled polymers?”

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### SESSION 2: Fracture of Heterogeneous Material

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**16:45**

*Jian Ping Gong, Hokkaido University (Japan)*

“Tough hydrogels based on sacrificial bond principle”

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**17:30**

*Stéphane Roux, ENS Cachan (France)*

“Watching cracks”

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**18:15**

End

## December 10th

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**09:00**

*Anthony Kinloch, Imperial College London (UK)*

“The mechanics and mechanisms of fracture of nanomodified epoxy polymers”

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**09:45**

*Herbert Hui, Cornell University (USA)*

“Effect of large strains on crack tip fields in nonlinear elastic solids”

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**10:30**

Coffee Break

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### SESSION 3: Adhesion and Friction at Heterogeneous interfaces

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**11:00**

Keynote: *Christian Frétiigny, Antoine Chateauminois, Yohan Le Chenadec, ESPCI ParisTech & Michelin (France)*

“Local friction of rubber with rough surfaces”

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**11:30**

*Étienne Barthel, CNRS–Saint-Gobain (France)*

“Can we engineer fracture and wetting properties through interface textures?

An experimental viewpoint on elastic line pinning”

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**12:15**

Lunch break: Buffet

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**13:45**

*Jay Fineberg, The Hebrew University of Jerusalem (Israel)*

“How Things Slide: Rapid Dynamics at the Onset of Friction”

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**14:30**

*Eduard Arzt, INM- Leibniz-Institut für Neue Materialien (Germany)*

“Bioinspired adhesive systems: from ideas to applications”

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**15:15**

*Walter Federle, University of Cambridge (UK)*

“Wet but not slippery: how insects climb on plant surfaces”

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**16:00**

Conclusions: *Maude Portigliatti (Michelin)*

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**16:30**

Coffee; end of workshop

## Invited Speakers

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**Eduard Arzt**

Leibniz-Institut für Neue Materialien gGmbH  
66123 Saarbrücken, Germany

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**Étienne Barthel**

Surface du Verre et Interfaces  
CNRS/Saint-Gobain  
93303 Aubervilliers, France

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**Jay Fineberg**

The Racah Institute of Physics  
The Hebrew University of Jerusalem  
Givat Ram, Jerusalem 91904, Israel

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**Jian Ping Gong**

Laboratory of Soft & Wet Matter  
Faculty of Advanced Life Science, Hokkaido University  
Sapporo 060-0810, Japan

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**Herbert Hui**

Mechanical and Aerospace Engineering  
Cornell University  
322 Thurston Hall Ithaca, New York 14853, USA

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**Anthony Kinloch**

Department of Mechanical Engineering  
Imperial College London  
South Kensington Campus, London SW7 2AZ, UK

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**Gareth H. McKinley**

Department of Mechanical Engineering,  
Massachusetts Institute of Technology  
Cambridge, MA 02139, USA

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**Peter Olmsted**

Department of Physics & Astronomy  
University of Leeds,  
Leeds, LS2 9JT, UK

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**David Pine**

Center for Soft Matter Research  
New York University  
4 Washington Place, New York, NY 10003, USA

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**Olivier Pouliquen**

IUSTI, Polytech Marseille  
5, rue Enrico-Fermi, 13453 Marseille, France

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**Stéphane Roux**

L.M.T. Cachan  
61, avenue du Président-Wilson, 94235 Cachan, France

## Poster Abstracts

P1

Polymer and Interfaces:

Atomistic simulations and continuum approaches

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**Mathieu Solar**

*Martin-Luther-Universität Halle-Wittenberg, Germany*

[mathieu.solar@physik.uni-halle.de](mailto:mathieu.solar@physik.uni-halle.de)

The appropriate modeling of matter is becoming an increasingly unavoidable step in the predictive numerical simulations of phenomena like adhesion/adherence taking part of indentation (or scratch) tests and cracking or damage of polymers. In such modeling, the matter is considered from two points of view: a continuum and an atomistic point of view. This poster presentation illustrates former works on the rheology of polymer melts and films in the glassy and near the

rubbery domain using two different methods: molecular dynamics (MD) and finite element (FE) simulations. Some comparisons between both methods are proposed and our results provided evidence in favor of using MD simulations to investigate the physics of polymer interfaces, since the volume elements studied were representative and thus contained enough information about the microstructure of the polymer model.

## P2

### Structure and Dynamics at Polymer-solid interfaces: MD simulations of 1,4 - polybutadiene at graphite surfaces

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#### Mathieu Solar

*Martin-Luther-Universität Halle-Wittenberg, Germany*

[mathieu.solar@physik.uni-halle.de](mailto:mathieu.solar@physik.uni-halle.de)

Polymer dynamics in confinement is both of fundamental interest concerning our understanding of the glass transition, as well as of high technological importance for the performance of composite materials. The results here presented are concerned with atomistic MD simulations of a chemically realistic model of a 1,4-polybutadiene melt (55% trans and 45% cis content, randomly dispersed) confined between two walls of graphite. The focus of our study is to

investigate the effects of confinement on the chain dynamics in the melt and to reveal to what extent the walls are influencing structure and dynamics of the melt. The physical properties here investigated are concerned with the structure and the topology of the chains in the confined melt and also with dynamic relaxation of various chain motions. The results presented are the key to a better understanding of the glass transition process in a confined polymer system.

## P3

### Mechanical reinforcement in confined polymers(\*)

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#### Alexey V. Lyulin

(presented by Theodoros Davris)

*Eindhoven University of Technology, The Netherlands*

[a.v.lyulin@tue.nl](mailto:a.v.lyulin@tue.nl)

[t.davris@tue.nl](mailto:t.davris@tue.nl)

Constant-pressure NpT molecular-dynamics computer simulations have been carried out for a bead-rod polymer model confined between two attractive crystalline substrates. Different film thicknesses and different polymer-substrate interaction strengths were used. The density profiles for simulated films show a monomer layering close to the polymer-substrate interface. Higher density was found in this region compare to the middle bulk-like film layers. It was shown that decreasing film thickness leads to an increase of density, as well as to the film-averaged glass transition temperature compare to the bulk. Layer-resolved analysis of the segmental dynamics of the thickest films showed a gradient of the mobility when approaching the polymer-substrate interface, while the middle-layer dynamics exhibits bulk-like behaviour. By decreasing film thickness, these gradients overlap. Polymer films were deformed uniaxially and their elastic moduli were extracted. The mechanical

reinforcement was found to increase with decreasing film thickness; this effect is explained by analyzing the layer-resolved relaxation times. For all films in the linear regime of deformation the secant modulus was found to fluctuate around an average value which is close to the film elastic modulus. In a non-linear regime a drop of the secant modulus is observed. This drop is larger in thinner films and weaker in the bulk, thus showing a decrease of the overall film reinforcement with deformation. At the same time, the layer-resolved orientational relaxation times were found to decrease upon increasing strain amplitude; the latter acceleration effect strongly depends on the distance from the substrates at different substrate-attraction strengths. It is shown that the drop of reinforcement can be qualitatively explained by the decrease of the relaxation times upon deformation, and by the heterogeneity of the orientational dynamics in case of ultra-thin polymer films.

(\*) *Chrysostomos Batistakis, M.A.J. Michels, and Alexey V. Lyulin*

P4

## Hybrid Magnetic Liquid Crystalline Materials

**Monique Mauzac**

Laboratoire IMRCP, CNRS, Toulouse, France

[mauzac@chimie.ups-tlse.fr](mailto:mauzac@chimie.ups-tlse.fr)

Organic-inorganic soft materials, responsive to a magnetic field, are increasingly studied as alternative to the older rigid magnetic networks usually characterised by low flexibility and small shape changes [1, 2]. At present, the main approach to obtain such materials consists in mixing an inorganic, field sensitive component (most frequently iron particles of nano- or microsized) and an organic deformable matrix. New properties, such as shape change (using an electro-magnetic induced heating on doped shape memory polyurethanes or polyacrylates [3, 4]) and tunable elastic modulus (doped polysiloxanes cross-linked in the presence of a magnetic field [5, 6]), for example, are aimed. In this context liquid crystal materials are a

very interesting matrix [7, 8]. Our approach consists in mixing preformed ferromagnetic nanoparticles inside a cross-linkable liquid crystal siloxane [9, 10]. We will report here the synthesis of the composites and their properties from the mesomorphous to the magnetic ones will be described by means of X-ray scattering, Transmission Electron Microscopy and magnetic measurements. The final objective is to combine the advantages of liquid crystal elastomers (good elastic properties, possibility to reversibly undergo very important shape changes, anisotropic behaviour over a temperature range) with the wireless action of a magnetic field.

P5

## Dilatancy in granular media under shear: A route to closer packing and to flow? (\*)

**Nicolas Rivier**

Institut de Physique et Chimie des Matériaux de Strasbourg & Université de Strasbourg, Strasbourg, France

[nick@fresnel.u-strasbg.fr](mailto:nick@fresnel.u-strasbg.fr)

The densest packing of a hard, dry granular material (modelled as hard spheres of equal radius and infinite tangential friction that forces the grains to roll without slip on each other, or to lose contact) is a geometric, many-body optimisation problem with solutions on the boundary of configuration space: Physical contacts between grains are struts of minimal length. The boundary is made of smooth regions (one grain rolling on another), separated by (Pareto) points where the grains are stuck. The optimisation problem consists therefore in identifying the Pareto points in an arbitrary, disordered packing, and in exploring physically (by shearing) the configuration space in order to reach another Pareto point with higher density. The apparent paradox of dilatancy (wet footprint in the sand) was pointed out by O. Reynolds in 1885. It is explained simply in a minimal model of the granular material («sand») as hard spheres with infinite tangential friction, packed together as tetrahedra (Delaunay tessellation, the dual of Voronoi's) and making up a graph. Upon external stress, the spheres can either roll on each other without slip - impossible in circuits of odd numbers of grains in contact -, or lose contact with each other. Uniaxial stress (the vertical foot's pressure) shears the granular material; it forces the grains to roll on each other and the odd circuits to open up. Each tetrahedron of spheres distorts to break the triangular circuits of grains in contact, thereby increasing its volume and letting in water that is expelled in the wet footprint left behind.

How do the tetrahedra pack together? Is the distortion transmitted consistently from one tetrahedron to the next? Can disorder be accommodated? To every packing of spheres corresponds a (Delaunay) tessellation into tetrahedra. The vertices are the spheres' centers, and the edges represent either physical (struts) or topological «contacts» of non-minimal length (disconnected neighbors). Densest packing minimizes the total volume of the tetrahedra. Under shear, each tetrahedron deforms into a polygon of four struts and two opposite and orthogonal topological edges (three possibilities). The deformation is consistent throughout the tessellation (a result proven by regarding the dual Voronoi foam as an Eulerian graph). The packing has then only even circuits of struts and each grain can thus roll on its neighbors without slip and explore freely the (boundary of the) configuration space. The Pareto points are readily identified as local, cubic or hexagonal close-packed configurations (Kepler-Hales), as are the simplest non-Pareto configurations. Shear can induce elementary local topological transformations that may result in closer packing (topological annealing).

Notably, five tetrahedra sharing an edge constitute a defect that becomes, under further shear (and loss of contact of the two grains on the common edge), a dislocation that can glide away. This pentagonal dipyrmaid - a ten-faced deltahedron - is the essential nearly close-packed elementary topological configuration that can be annealed away.

(\*) *Nicolas Rivier and Jean Yves Fortin*

P6

## Toughness and Ductility of microporous carbons from molecular simulation

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**Laurent Brochard**

*Laboratoire Navier (CNRS/ENPC/IFSTTAR), Université Paris-Est, France*

[laurent.brochard@enpc.fr](mailto:laurent.brochard@enpc.fr)

We propose an approach by molecular simulation to estimate fracture properties of materials by molecular simulation. This approach is based on a thermodynamic integration and is valid whatever the mechanical behavior of materials. Therefore, it applies both to ductile and brittle materials. The methodology is validated by considering silica for which experimental data are available and the classical theory of linear

elastic fracture mechanics applies. Then, we consider a microporous carbon and its interface with silica. Both the bulk material and the interface exhibit a significant ductility. This microporous carbon is a molecular analogue of kerogen, the organic matter in shale. This work is a first step toward a fine understanding of the fracture properties of organic-rich shales.

P7

## Fingering to fracturing transition in a transient gel

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**Guillaume Foyart**

*Laboratoire Charles Coulomb, Montpellier, France*

[guillaume.foyart@univ-montp2.fr](mailto:guillaume.foyart@univ-montp2.fr)

Fracture processes are ubiquitous in soft materials, even in complex fluids, subjected to stresses. To investigate these processes in a simple geometry, we use a model self-assembled transient gel and study the instability patterns obtained in a radial Hele-Shaw cell when a low viscosity oil pushes the more viscous transient gel. Thanks to an analysis of the morphology of the patterns, we find a discontinuous transition between the standard Saffman-Taylor fingering instability and a fracturing instability as the oil injection rate increases. Our data suggest that the flow properties of the gel ahead of the finger tip control the transition towards fracturing. By analyzing the

displacement field of the gel in the vicinity of the fingers and cracks, we show that in the fingering regime, the oil gently pushes the gel, whereas in the fracturing regime, the crack tears apart the gel, resulting in a strong drop of the gel velocity ahead of the crack tip as compared to the tip velocity. We find a unique behavior for the whole displacement field of a gel around a crack, which is drastically different from that around a finger, and reveals the solid-like behavior of the gel at short time. Our experiments and analysis provide quantitative yet simple tools to unambiguously discriminate a finger from a crack in a visco-elastic material.

P8

## Development of optimized batch sSBR functionalized structures for low RR applications

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**Antonio Giuseppe Solito**

*Versalis S.p.A., Italy*

[antonio.solito@versalis.eni.com](mailto:antonio.solito@versalis.eni.com)

In the field of tire manufacturing the need of highly engineered materials has recently become crucial to face up to the increasingly stricter requirements dictated by the EU regulations on the subject of fuel consumption, safety and noise levels. For this reason, in the last few years, a lot of efforts have been made to fulfill these requirements either working on the synthesis of sSBRs with optimized molecular architecture and chemical composition or developing chemically modified structures. As far as the molecular architecture is concerned, it has been shown that batch polymerized sSBRs are good candidates for low rolling resistance (RR) compared to continuous sSBRs mainly due to a narrower molecular weight distribution and the absence of low molecular-weight fractions responsible for hysteresis phenomena. Nevertheless, as widely described in the literature, chain-end functionalization is an effective alternative to reduce RR and to enhance tire performance without compromising processability during mixing. It is important to notice that the number of chain-ends available before functionalization depends on both, the polymer molecular weight and, in the case of star-coupled structures, the coupling efficiency (i.e. the weight fraction of star-shaped molecules). The higher the molecular

weight, the lower is the number of chain-ends and vice versa. Furthermore, when a coupling agent is used, the number of chain-ends available for functionalization decreases even more. It follows that the most advantageous performance in terms of RR can be obtained predicting the trade-off between molecular architecture (molecular weight and coupling efficiency) and number of functionalized chain-ends with respect to this parameter. In this work different polymeric sSBR blends have been synthesized using a tetrafunctional coupling agent. For each blend, variations of coupling efficiency and molecular weight of the uncoupled fraction have been made. Moreover, for each structure different kinds of functionalizing agents have been tested. For the resulting matrix of grades obtained, an extensive chemical, physical and technological characterization has been carried out to investigate the behavior of both, the raw polymers and their silica-based vulcanized compounds. As a result, important trends have been extrapolated and allowed the individuation of a target macrostructure capable of significantly enhancing the effect of terminal functionalizing groups bound to the chain-ends of the uncoupled linear polymer fraction.

P9

## A mesoscopic description of the shear flow of athermal amorphous solids

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**Alexandre Nicolas**

*Université de Grenoble-Alpes, Grenoble, France*

[alexandre.nicolas@ujf-grenoble.fr](mailto:alexandre.nicolas@ujf-grenoble.fr)

In spite of their extreme diversity, amorphous solids, ranging from foams and jammed emulsions to molecular glasses, display a similar phenomenology when they are forced to flow: on top of a basic elastic response, the material features localised regions where particles rearrange rapidly; these local shear transformations, also known as plastic events, induce long-range elastic deformation field. We develop a mesoscopic model directly based on this simple scenario. In particular, we focus on situations where

thermal fluctuations are negligible, and show that the model captures the peculiarities of the flow of these materials. Besides the macroscopic properties, we consider the spatial correlations between plastic events. Because the long-range elastic deformations are properly taken into account in the model, these correlations are found to be in good agreement, at least qualitatively, with results from atomistic simulations.

P10

## Properties of nanoparticle ribbons prepared by flow coating

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**Jonathan Pham**

*University of Massachusetts Amherst, USA*

[jpham0@polysci.umass.edu](mailto:jpham0@polysci.umass.edu)

Materials that possess structural hierarchy display a range of properties and multi-functionality across multiple length scales. Among such structures are nanoparticle assemblies, which have gained much interest for their potential use in a wide range of applications. We have developed a technique that utilizes a geometrically confined coffee ring effect, called flow coating, to direct the assembly of nanoparticles into ribbons and grids. The assemblies are constructed with photoreactive quantum dots and crosslinked by UV irradiation affording long, flexible and robust structures that allow for subsequent

liftoff from the substrate. The structural integrity of freely floating grids and high aspect ratio ribbons are observed through fluorescence microscopy. We find that these nanoparticle assemblies possess unique properties such as two-dimensional flexibility and the self-formation of helical structures. Helical structures are formed through the balance between interfacial tension and elasticity of the geometrically asymmetric ribbons. Nanoparticle helices are shown to have excellent structural integrity with spring-like characteristics and can be extended to high strains.

P11

## Large deformation and fracture of highly deformable gels having permanent and transient crosslinks

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**Koichi Mayumi**

*ESPCI Paristech, Paris, France*

[koichi.mayumi@espci.fr](mailto:koichi.mayumi@espci.fr)

We have studied the dissipative behavior of highly deformable dual crosslink gels having covalent crosslinks and reversible crosslinks with a well identified life time. The breakage of transient crosslinks dissipates strain energy and enhances the fracture toughness and extensibility of the dual crosslink gels. We performed systematic studies on large strain mechanical properties of the dual crosslink gels over 4 orders of magnitudes of strain rates and proposed a

simple equation to describe their stress-strain curves. The equation is composed of strain and time dependent terms. The strain dependent term is predicted by the classical rubber elasticity theory and the time dependent term is attributed to the associative Rouse mode. Moreover, we conducted fracture tests on the dual crosslink gels. The fracture energy of dual crosslink gels is ten times as large as that of chemical gels without physical crosslinks.

P12

## Evidence for a unique structure-determining parameter in simplified industrial styrene-butadiene/silica nanocomposites

**Guilhem Baeza**

*Laboratoire Charles Coulomb and Michelin, Montpellier, France*

[guilhem.baeza@univ-montp2.fr](mailto:guilhem.baeza@univ-montp2.fr)

Silica-filled styrene-butadiene nanocomposites with grafted polymer molecules have been formulated for various silica volume fractions ( $\approx 10\%$  and  $20\%$ ), chains masses ( $40 - 280 \text{ kg.mol}^{-1}$ ), and fractions of graftable chains ( $0 - 100\%$ ). The silica structure was analyzed using electron microscopy and small-angle x-ray scattering. The nominal density of grafted molecules per unit silica surface, which depends on all three aforementioned parameters, covered a domain from 0 to about  $0.1 \text{ nm}^{-2}$ . By comparing the filler structures at fixed grafting density (so-called 'twins'),

a surprising match of the microstructures was evidenced. A quantitative tool expressing the similarity of structures in reciprocal space is proposed. By comparing it to typical structural evolutions of this nanocomposite system, the similarity is unambiguously proven. Dynamical mechanical analysis was used to compare the mechanical properties of the twins. The elastic plateau modulus is found to be identical for twins, highlighting the possibility to tune the flow behavior of nanocomposites at fixed structure and modulus via the chain length at constant grafting density.

P13

## Rheological behaviour of Styrene-Butadiene Rubber at low temperature, pure and filled with carbon black, and application to extrusion flows(\*)

**Rudy Valette**

*CEMEF, MINES ParisTech, Sophia Antipolis, France*

[rudy.valette@mines-paristech.fr](mailto:rudy.valette@mines-paristech.fr)

The flow behaviour of a synthetic SBR rubber is studied by capillary rheometry in the range of temperatures encountered in extrusion, i.e. between  $90^\circ\text{C}$  and  $40^\circ\text{C}$ . A pure SBR and SBR compounds filled with carbon black (with different amounts, from 16 to 33 wt%) are characterized. We observed, for all tested materials, that the flow curve can be divided in different parts: at low shear rate, the classical behaviour of a polymer is evidenced, with the stress increasing regularly with the shear rate. Above a certain critical stress, flow conditions change, with the onset of wall slip and/or upstream instabilities. The critical stress is independent of the temperature but increases linearly

with the carbon black amount. The flow curves at different filler content can be superimposed, using a shift factor depending on filler content. We can thus propose a general viscosity law, based on a Carreau-Yasuda behaviour for which zero-shear viscosity and characteristic time depend on both temperature and filler content, through Arrhenius and homogenized Krieger-Dougherty expressions, respectively.

Application to extrusion flows will be shown through a comparison between experimental measurements and 3D direct flow modelling based on the obtained rheological model. The role of wall slip in the model predictions will be discussed.

(\*) A. Cri 1,2, C. Baritaud2, V. P lissier2, R. Valette1, B. Vergnes1

1 MINES ParisTech, CEMEF, UMR CNRS 7635, BP 207, 06904 Sophia Antipolis (France)

2 Michelin, Centre de Technologie Europe, 63040 Clermont Ferrand (France)

P14

## Ultrasonic imaging of carbon black gels under oscillatory shear

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**Sébastien Manneville**

*Laboratoire de Physique de l'ENS-Lyon, Lyon, France*

[sebastien.manneville@ens-lyon.fr](mailto:sebastien.manneville@ens-lyon.fr)

Attractive colloidal gels display a solid-to-fluid transition as shear stresses above the yield stress are applied. This shear-induced transition is involved in virtually any application of colloidal gels. It is also crucial for controlling material properties. Still the yielding transition is far from fully understood. For instance we find that gels stressed below the apparent yield stress inferred from small-amplitude oscillatory shear can eventually flow if given enough time. Moreover classical rheology experiments do not provide any local insight on such «delayed yielding». In particular, the loss modulus may dominate the storage modulus

only because the sample has yielded close to a wall while it remains solid-like in the bulk. To go beyond simple rheology experiments we use a high-frequency ultrasonic scanner to image the local displacements of opaque attractive colloidal gels (carbon black gels) under large amplitude oscillatory shear. We find that yielding is both heterogeneous in time and space and we isolate various yielding mechanisms as a function of the amplitude and frequency of the applied oscillatory stress. The amplitude dependence of the yielding behavior hints toward activated processes in line with recent models.

P15

## Rheo-ultrasonic imaging of secondary flows in a Taylor-Couette device

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**Marc Fardin**

*Laboratoire de Physique de l'ENS-Lyon, Lyon, France*

[marcantoine.fardin@ens-lyon.fr](mailto:marcantoine.fardin@ens-lyon.fr)

The Taylor-Couette device is a geometry with two concentric cylinders, which is commonly mounted on commercial rheometers and is used extensively to study the material properties of a variety of complex fluids. When the gap between the cylinders is small the base azimuthal flow is viscometric, but if the base flow becomes unstable, secondary flows emerge and break the viscometric assumption. In particular, in a Newtonian fluid inertia generates Taylor vortices. Similar secondary flows are also observed in polymer or wormlike micelles solutions due to a viscoelastic

instability. In all cases, secondary flows deform the azimuthal base flow, which loses its axial invariance. We report on a new ultrasonic imaging technique that allows to follow such deformation of the main flow by measuring the time-resolved azimuthal velocity profiles all along the axis of the cylinders simultaneously to the rheological response. We show examples of the dynamics of secondary flows for several Newtonian and non-Newtonian fluids and we stress their impact on viscometry.

P16

## Shear-induced structures and flow instabilities

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**Marc Fardin**

*Laboratoire de Physique de l'ENS-Lyon, Lyon, France*

[marcantoine.fardin@ens-lyon.fr](mailto:marcantoine.fardin@ens-lyon.fr)

We study the Taylor-Couette flow of a dilute micellar system known to generate shear-induced structures. We show that flow instabilities must be taken into account since both the Reynolds number and the Weissenberg number may be large. Before nucleation of shear-induced structures, the flow can be inertially

unstable, but once shear-induced structures are nucleated the kinematics of the flow become chaotic, in a pattern reminiscent of the inertio-elastic turbulence known in dilute polymer solutions. We outline a general framework for the interplay between flow instabilities and flow-induced structures.

P17

## Elastic instabilities in the flow of viscoelastic fluids

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**Laura Casanellas**

*Université Paris-Diderot and ESPCI ParisTech, Paris, France*

[lauracasanellasvilageliu@gmail.com](mailto:lauracasanellasvilageliu@gmail.com)

Viscoelastic fluids exhibit an intermediate behavior between elastic solids and dissipative liquids, depending on the time scale at which they are perturbed. Elastic instabilities may occur in the flow of viscoelastic fluids and a general criterion for the onset of such instabilities can be established based on the

curvature of flow streamlines and fluid elasticity. Our goal is to study the onset of elastic flow instabilities in viscoelastic fluids (polymeric fluids and wormlike micellar solutions) of various rheologies in different flow geometries, from macro to microfluidic devices.

(\*) *Laura Casanellas, Anke Lindner, Sandra Lerouge*

P18

## Surface tension between a colloidal solution and its solvent

**Domenico Truzzolillo**

*Laboratoire Charles Coulomb - Université Montpellier 2, Montpellier France*

[domenico.truzzolillo@univ-montp2.fr](mailto:domenico.truzzolillo@univ-montp2.fr)

The intrusion under pressure of a low viscosity fluid into another high viscosity phase may lead to viscous fingering: an hydrodynamic instability whose onset and time evolution have been widely studied since 50s. The initial development of the interface instability tracks the predictions of the linear stability theory that, hence, is a suitable tool to predict the onset of fingering. Nevertheless a closed form for the observable wavelength of the perturbation at the onset of the instability was still lacking. We show that the growing

unstable mode with the maximum amplitude at the onset of the Saffman-Taylor instability predicted by the linear evolution theory is the correct observable related to the number of fingers. The knowledge of the single particle dimension  $d$  and the flow properties of the solutions at different polymer volume fractions  $\Phi$  allows to indirectly infer the values of the interfacial tension  $\Gamma_e$  between two miscible phases. We found  $\Gamma_e = \kappa \Phi^2 / d$  in agreement with the square gradient model prediction.

P19

## Wrinkling and yielding of casein gels

**Mathieu Leocmach**

*Laboratoire de Physique de l'ENS-Lyon, Lyon, France*

[mathieu.leocmach@ens-lyon.fr](mailto:mathieu.leocmach@ens-lyon.fr)

We report on nested wrinkling patterns of protein gels, which spontaneously emerge during the gelation of a protein solution in a confined space. The slow acidification of a sodium caseinate solution leads to the formation of a gel. Practical acidification procedures, either biological (yeast) or chemical (acids with slow opening kinetics, here delta-gluconolactone) often over-acidify the solution below the isoelectric point of the protein. The shear modulus of the gel thus drops after the initial gelation. Here, we use non-adhesive boundary conditions on the top and bottom

of our cell to translate the shear modulus overshoot into a volume overshoot. This procedure leads to an initial shrinkage of the gel (syneresis) into a film thinner than the cell, followed by dilation and wrinkling. The amplitude of the wrinkles being constrained by the volume of the cell, nested wrinkles are generated. We make a parallel between this self-induced wrinkling under non-adhesive boundary conditions and the fractures observed in creep experiments on the same system under adhesive boundary conditions.

## P20

### Entanglement-induced reinforcement in polymer nanocomposites

**Samy Merabia**

CNRS and Université Lyon 1, Lyon, France

[samy.merabia@univ-lyon1.fr](mailto:samy.merabia@univ-lyon1.fr)

We propose a coarse-grained model able to describe filled entangled polymer melts. Our purpose is to study the reinforcement caused by the effect of fillers on the entanglement network, as speculated in previous experimental work, and also observed in molecular dynamics studies. In this work, the filler volume fraction effect, the distribution of the fillers (cubic lattice, randomly dispersed, and small clusters randomly dispersed) and the presence or absence of grafted chains on the fillers are investigated. Our model is based on a “slip-link” model initially developed to study the entanglements in pure polymer melts and offers a less costly computational method than molecular dynamics simulations for the study of entangled polymer melts. The polymer chains are described as Rouse chains of Brownian particles connected by Hookean springs, and are subject to friction and random forces. Entanglements are artificially imposed by objects (slip-links) exhibiting statistical fluctuations that do not modify the equilibrium statistics of the melt. In addition we introduced excluded volume interactions between chain segments, to take into account the incompressibility of the melt.

These excluded volume interactions do not perturb the dynamics of the chains in the homogeneous limit as expected from theoretical considerations on short range interactions. Finally, the fillers are modeled by immobile spherical objects, with or without grafted chains, which interact with a repulsive potential with the chain monomers. The chains grafted onto the fillers are represented by “additional slip links” confined in the vicinity of each filler. We first present the effect of the filler distribution and filler volume fraction, considering only bare fillers. Then, the effect of grafted chains via the additional slip-links is also shown as a function of the same parameters. Our results show that the presence of grafted chains induces an important change in the viscosity, calculated by integrating the stress autocorrelation function. Both the plateau value and the terminal relaxation time depend on the density of fillers and on the number of grafted chains. Moreover, we find that a disordered filler configuration induces confinement effects that amplify reinforcement compared to the case of a perfectly ordered configuration.

[1] D. Del Biondo, E. Masnada, S. Merabia, M. Couty and J.-L. Barrat, *J. Chem. Phys.* 2013

[2] E. Masnada, S. Merabia, M. Couty and J.-L. Barrat, *Soft Matter* 2013

## P21

### Heterogeneous molecular dynamics of thin polyisoprene film between silica surfaces

**Daria Guseva**

Eindhoven University of Technology, Department of Applied Physics, The Netherlands

[d.v.guseva@tue.nl](mailto:d.v.guseva@tue.nl)

Mechanical properties of elastomer-based nanocomposite materials may be improved by adding filler particles. However, the connection of structure and dynamics of filled elastomers to the chemical constitution and interactions of the macromolecules and nanoparticles of which they are composed is still not fully understood and requires further investigation. In this paper, constant-pressure NpT molecular-dynamics computer simulations have been carried out for united-atom model of a non-crosslinked cis-1,4 polyisoprene film confined between two amorphous, fully-coordinated silica surfaces. Film thickness  $D$  has been varied in a wide range,  $1 < D/R_g < 9$ , where  $R_g$  is the individual chain radius of gyration. After a thorough equilibration, the film stratified structure and dynamic properties (layer density of polyisoprene, orientational relaxation autocorrelation functions of

chain bond vectors) at different temperatures have been studied. Both structure and segmental dynamics in the middle of the films resembles that in a corresponding bulk. Polymer density profiles show pronounced ordering of polymer segments close to the vicinity of silica, this ordering disappears towards the film middle. Monomers in thin films are found to be oriented along the silica plane. Tremendous slowing down of polymer segmental dynamics relaxation has been observed in the film surface layers, 170 times slower as compared to that in a bulk. This effect increases with decreasing the film thickness and temperature. As a consequence, decreasing the confinement film thickness results in an increase of averaged  $T_g$  in films. The interpretation of the results in terms of the polymer glassy bridges has been discussed.

P22

## Large-scale cg-MD simulation of polymer nano-composite filled with nano-particles

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**Hagita Katsumi**

*National Defense Academy of Japan, Japan*

[hagita@nda.ac.jp](mailto:hagita@nda.ac.jp)

Coarse-grained Molecular Dynamics (MD) of Kremer-Grest model is powerful tool to study mechanical properties of multicomponent soft materials. As a challenge of multiscale / multiphysics simulation, we extended applications of this type of simulation

method from polymer melts to filled polymer (rubber) nano-composites. We will present a brief review of simulation studies of polymer rubber materials filled with nano particles, whose morphology is modelled from Ultra Small Angle X-ray Scattering data.

P23

## Avalanche Dynamics in Sheared Granular Material

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**Ramon Planet Latorre**

*Institut Lumière Matière, Université de Lyon, 69622 Villeurbanne, France*

[ramon.planet-latorre@univ-lyon1.fr](mailto:ramon.planet-latorre@univ-lyon1.fr)

We present an experimental study of the velocity fluctuations of a granular medium under constant shear. Our setup shows that the system display an intermittent behavior signature of a burstlike dynamics, with power-law distributed avalanches.

Acoustic, visual and force measurements are recorded with two main objectives: i) to study the role of disorder on the criticality of a system; ii) to study the prediction of catastrophic events in scale invariant avalanches

P24

## Scale invariant avalanches: a critical confusion

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**Ramos Osvanny**

*Institut Lumière Matière, Université de Lyon, 69622 Villeurbanne, France*

[osvanny.amos@univ-lyon1.fr](mailto:osvanny.amos@univ-lyon1.fr)

In the last decades, considerable efforts have been devoted to understanding single events related to friction, fracture and unjamming transition, commonly denominated avalanches. However, in many different natural scenarios -from subcritical fracture to earthquake dynamics- these events are of all scales; a situation that has often been interpreted within the formalism of critical phenomena, and having as a relevant consequence the inherently unpredictability of

scale-invariant avalanches. A revision of this interpretation which departs from standard ideas is presented here, resulting in: (i) critical systems are not necessarily unpredictable; (ii) slowly driven systems evolving through power-law distributed avalanches are not necessarily critical; and (iii) scale-invariant avalanches are not necessarily unpredictable. Simple simulations and granular experiments confirm the findings.

P25

## Revealing the structure of a granular medium through acoustic measurements

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**Sébastien Lherminier**

*Institut Louis Lumière/ENS Lyon, Lyon, France*

[sebastien.lherminier@ens-lyon.fr](mailto:sebastien.lherminier@ens-lyon.fr)

An array of acoustic sensors records the sound that has travelled across a bi-dimensional granular medium, consisting of photoelastic discs, which are confined between two transparent plates and arranged into different crystalline or disordered structures. The system is compressed along one direction (either force-controlled or displacement-controlled) and can be sheared in the direction perpendicular to the ap-

plied force; while the acoustic signals are generated through a well-controlled and local mechanical excitation. The results show power-law regimes in the force vs. sound speed relation, with exponents that are sensitive to the structure of the packing. Small structural changes are also detectable which, in principle, can be used to predict large avalanches during the slow shearing of the system.

P26

Acoustic emission in fracturing paper:  
Statistics and correlation effects

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**Menka Stojanova**

*Institut Lumière Matière, Université de Lyon, 69622 Villeurbanne, France*  
[menka.stojanova@univ-lyon1.fr](mailto:menka.stojanova@univ-lyon1.fr)

The subcritical propagation of a fracture in a sheet of paper is observed using two methods: low frequency visualization and high frequency acoustic monitoring. The crack advances through a series of discrete fracture events. Though it seems impossible to match acoustic and visual events individually, similar global activities have been observed using both methods. Furthermore, they result in power law distributions of waiting times between successive events with the same exponents. However, the distributions

of events' energy follow power laws with different exponents. We show that this difference is related to the different frequencies of the observation methods: by lowering the frequency of analysis of the acoustic data to the images' frame rate, similar exponents are obtained for both energy distributions. This is resulting from the existence of time correlations between the events, in particular aftershocks, which are detected by the acoustics but not by the image analysis.

P27

Stick-slip crack growth instability in adhesive tapes

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**Marie-Julie Dalbe**

*Laboratoire de Physique de l'ENS-Lyon, Lyon, France*  
[mariejulie.dalbe@ens-lyon.fr](mailto:mariejulie.dalbe@ens-lyon.fr)

We study experimentally the fracture dynamics during the peeling at a constant velocity of an adhesive tape. An original peeling setup which we have developed allows to peel the adhesive from a plane substrate while setting the mean peeling angle to a constant value. This experimental achievement is made possible by the coupled motions of the substrate and of the peeled tape end at identical velocities thanks to two electronically enslaved motors. Thanks to a high speed camera, we measure, in an intermediate range of peeling velocities, high frequency oscillations between phases of slow and rapid propagation of the peeling fracture. This so-called stick-slip regime is well known as the consequence of decreasing fracture energy of the adhesive in a certain range of peeling velocity coupled to the elasticity of the peeled tape. We

are able to access directly the peeling point dynamics and to study quantitatively the Stick-Slip features (Stick and Slip periods and velocities) as a function of the three control parameters which are relevant to the problem: the mean peeled tape length, the mean peeling velocity and peeling angle. We report various Stick-Slip peeling regimes depending on the imposed peeling velocity with periods of Stick and Slip either independent or proportional to  $V$ .

In these experiments, controlling the peeling angle is a major improvement to understand the physics of adhesive peeling which is confirmed to be strongly dependent on the peeling angle, especially in the Stick-Slip regime. This general feature questions the validity of the usually admitted independence with the peeling angle of the fracture energy of adhesives.

P28

## In-situ study of the fragmentation process of agglomerated filler in a rubber matrix

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**Edith Peuvre-Disdier**

MINES ParisTech – CEMEF, Sophia Antipolis, France

[edith.disdier@mines-paristech.fr](mailto:edith.disdier@mines-paristech.fr)

We developed a transparent counter-rotating shear cell coupled with an optical microscope. The counter-rotating geometry allows us to stabilize the particle in suspension in a liquid matrix submitted to a shear flow relatively to the laboratory framework. It is then possible to observe the behaviour of this particle under the effect of hydrodynamics stresses. If the particle is an agglomerated object and if the hydrodynamics stresses are larger than its cohesion strength, fragmentation takes place. We thus applied this technique to study the fragmentation process of agglomerated fillers such as carbon black and silica

suspended in a styrene-butadiene rubber. Different fragmentation processes were observed and characterized. Quantities such as the minimum shear stress to be applied to reduce the size of the agglomerated filler are determined. The comparison of different grades of carbon black and precipitated micropearl silica allows us to better understand the mechanism of fragmentation of such agglomerated fillers. Conditions for fragmentation could be related to the agglomerate internal organisation. The experimental procedure, the validity of the measured data and the models used to interpret the data will be discussed.

P29

## LAOS: The strain softening/strain hardening paradox

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**Marius Mermet-Guyennet**

*Van der Waals-Zeeman Instituut, University of Amsterdam, The Netherlands*

[maris.mermet@gmail.com](mailto:maris.mermet@gmail.com)

Numerous materials, from biopolymers to filled rubbers, exhibit strain softening at high strains during a strain sweep in oscillatory rheology: the modulus decreases with increasing deformation. On the other hand, if the non-linear elastic response is analyzed within a single oscillation cycle (described by Lissajous curves), these systems are reported to exhibit strain hardening. We compare strain sweeps and single cycle LAOS (Large Amplitude Oscillatory Shear) analyses

of stress vs. strain on three very different materials. We conclude that the reported strain hardening is due to the use of a tangent modulus in the LAOS analysis, and that the overall rheology remains strain softening. To show that this conclusion is robust, we demonstrate a rescaling of the modulus that collapses the data from all the oscillatory measurements onto a single master curve that clearly exhibits the correct strain softening behavior.

## P.30

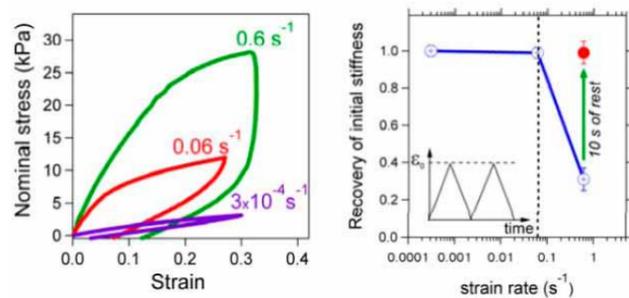
### Dissipation, fracture and recovery processes of Nano-Hybrid Hydrogels (\*)

**Alba Marcellan**

ESPCI ParisTech, Paris, France

[alba.marcellan@espci.fr](mailto:alba.marcellan@espci.fr)

The simplest sketch of a synthetic hydrogel consists of a 3D organic network, using hydrophilic polymer chains with chemical cross-linker to make inter-chains covalent bonds. Evidently, the resulting macromolecular architecture is a swollen material that mechanically performs poorly, that combines a very low modulus with high fragility. Over the past ten years, research has evolved from the simple idealization of a classical chemical gel, to envisaging diverse designs for tough gels, i.e. introducing additional dissipative mechanisms at the molecular level.



We developed new type of hybrid gels. The concept relies on combining a covalent network with physical cross-links by adsorption of polymer at inorganic particle surface. Such gels imply a rather simple and versatile chemistry by copolymerization of monomer with chemical cross-linker in the presence of inorganic nanoparticle suspension (water). The first results of these so-called nano-hybrid hydrogels highlighted a very unusual combination of properties [1]: elasticity, strength, dissipation and strain at failure were seen to be simultaneously enhanced by silica nanoparticle addition.

Fig.1: (a) Illustration of strain rate effect on hybrid hydrogel2: loading-unloading cycles for varied strain rates (from  $3.10^{-4} \text{ s}^{-1}$  to  $0.6 \text{ s}^{-1}$ ) on the same gel. (b) Time-dependence of recovery. Recovery is defined as the ratio between the initial modulus during the second loading cycle and the initial modulus during the first loading cycle. Only 10 seconds of rest enable the gel to recover its initial stiffness.

The characteristic times involved in the nanoparticle/polymer association were investigated by large strain mechanical cycling varying the strain rate [2]. The mechanical behaviour of the hybrid hydrogel was observed to vary tremendously over a relatively small range of strain rates, ranging from almost non dissipative (at slow strain rates) to highly dissipative at high strain rates. However, upon cycling over time-scales of tens of seconds, the physical associations taking place between nanoparticles and the polymer

network chains enabled the hydrogel to recover its initial mechanical properties. The main feature of this work is the remarkable role played by nanoparticles in the network to promote transient and recoverable connectivity by reversible adsorption/desorption processes. The strong strain rate dependence suggests that toughening mechanisms operating at standard strain rates, as often reported, maybe quite different at slower or larger strain rates.

[1] Carlsson, Rose, Hourdet, Marcellan, *Soft Matter* 2010

[2] Rose, Dizeux, Narita, Hourdet, Marcellan, *Macromolecules* 2013

(\*) Séverine Rose<sup>1</sup>, Tetsuharu Naritaz<sup>1</sup>, Dominique Hourdet<sup>1</sup>, Alba Marcellan<sup>1,2</sup>

1. Science et Ingénierie de la Matière Molle UMR 7615 (UPMC – CNRS – ESPCI), 10 rue Vauquelin, 75231 Paris Cedex 05, France.

2. Matière Molle et Chimie UMR 7167 (CNRS – ESPCI) / Université Pierre et Marie Curie, 10 rue Vauquelin, 75231 Paris Cedex 05, France.

## P.31

### Rheology of associative polybutadienes (\*)

**Régis Schach**

Manufacture Française des Pneumatiques Michelin, Clermont Ferrand, France

[regis.schach@fr.michelin.com](mailto:regis.schach@fr.michelin.com)

In the framework of the tire tread band applications, a good compromise between the rheology of the uncured compound and the mechanical properties of the cured materials is expected. Low viscosity is required to ensure a good productivity in the plant, but if we only use the molecular weight of the polymer to set the viscosity, we will end up, after curing, with a material with a high concentration of dangling chains in the network. These dangling chains will dissipate energy due to the retraction relaxation process, which will increase rolling resistance and consequently fuel consumption.

The use of labile functions grafted to the polymer, which can be strongly associated at low temperature (20 – 60 °C, typical rolling temperature of a passenger car tire) and dissociated at high temperature (100 – 120°C, temperature of the industrial process) could be of great help to optimize the properties of the material in both temperature ranges. In this work we studied the rheology of high molecular weight (150 000 g/mol) statistically grafted polybutadienes and low molecular weight (20 000 g/mol) telechelic polybutadienes. Three different chemical functions, referred to A, B and C, with different association energies, were tested.

(\*) R. Schach, E. Fleury, R. Matmour, *Manufacture Française des Pneumatiques Michelin, Clermont Ferrand, France.*

P.32

AMUFISE:

A multi-scale, multi-physics approach for sustainable mobility (\*)

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**Daniel Berghezan**

*Manufacture Française des Pneumatiques Michelin, Clermont Ferrand, France*

[Daniel.Berghezan@fr.michelin.com](mailto:Daniel.Berghezan@fr.michelin.com)

AMUFISE is the name of a 2009–2012 multi-partner program which received financial support from the French National Research Agency (ANR). This project, supervised by Michelin, gathered together the “Mechanics Laboratory” of École Centrale de Lille (Prof. Mathias BRIEU), the “Soft Matter Science and Engineering Laboratory” of ESPCI Paris Tech (Prof. Costantino CRETON), and the “Mechanics and Technology Laboratory” of École Normale Supérieure de Cachan (Prof. Stéphane ROUX). Our global objective was to open new innovation or optimization ways in the field of the filled elastomers fatigue. The global investigation aimed at understanding the damaging mechanisms in the vicinity of the crack tip, going from multiscale experimental or numerical studies to

predictive methods for material conception.

AMUFISE was based on two main coupled approaches:

- Analysis of the crack tip mechanical fields: evolutionary constitution law development, implementation of the law in a finite element software, and comparison between the local calculated displacements fields and the ones obtained by digital image correlation.

- Elaboration of original techniques to characterize the near-crack tip damage. The investigation tools allow for the detection of open cavities and their anisotropic features, from nanometric to micrometric sizes.

We will focus on the striking results of this project.

P.33

Adhesion of Soft Materials at Short Contact Times (\*)

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**Chelsea S. Davis**

*Laboratory of Physics and Mechanics of Heterogeneous Media, ESPCI ParisTech, France*

[chelsea.davis@espci.fr](mailto:chelsea.davis@espci.fr)

In the tire manufacturing industry, two layers of uncrosslinked rubber are brought into contact and required to adhere to one another very rapidly. The scientific details governing the formation and adhesive strength of these short contacts are not well understood. Several studies have sought to characterize the cohesion of uncrosslinked viscoelastic polymers as a function of contact time. The general conclusions of these studies show an increase of the separation strength and the work of adhesion as the contact time increases. This phenomenon is attributed to (1) the increase in contact area as polymer relaxation (surface wetting) occurs as well as (2) the interpenetration of chain ends across the interface. Our current study seeks to more reliably quantify this dependence of adhesion on contact time by utilizing a custom-built testing apparatus which allows us to have unprecedented control over all aspects of a contact adhesion test (approach and retraction veloci-

ties, maximum contact pressure, and contact time). This device is able to implement contact times over several orders of magnitude ranging from 0.03s to 100s. We have investigated the contact time effects of three unique contact scenarios: elastomer-silica, styrene-butadiene rubber (SBR) – silica, and SBR-SBR. With these three samples, we can decouple the effects of intermolecular forces, wetting/contact area, and chain interpenetration respectively. Additionally, we have begun a preliminary study seeking a better understanding of the role that oligomeric “tackifiers” play in increasing the cohesion of uncrosslinked rubber at short contact times without impacting the material properties of the system. Our study has practical implications which will directly impact the development of future tire manufacturing techniques as well as other industries where rubber-rubber interfacial strength is required.

(\*) *Chelsea S. Davis, David Martina, Costantino Creton, Anke Lindner*

