HELLENIC SOCIETY
OF RHEOLOGY

Athens, Greece
June 27-29, 2004

Programme & Abstracts
HSR 2004
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Proceedings compiled by Evan Mitsoulis. Cover page designed by Evan Mitsoulis.
CHAIR’S MESSAGE

It is with great pleasure that we welcome all delegates and accompanying persons to the 4th Meeting of the Hellenic Society of Rheology, HSR 2004, in Athens, Greece. HSR meetings are rotated every 3 years among various locations around Greece and Cyprus. This is the 4th meeting since the foundation of the Society in 1996, when its 1st meeting was held in Nicosia, Cyprus. The 2nd meeting was held in 1998 in Herakleion, Crete, and the 3rd meeting in 2001 in Patras. Athens has been chosen for this meeting, as it is the capital of Greece, with the largest population and activities, preparing frantically for the 2004 Olympic Games.

The HSR 2004 Meeting draws scientists and engineers from 13 countries. The Conference includes 2 plenary and 31 oral lectures and 17 posters. The scientific papers will be presented under 6 major themes. We hope that this Conference will promote and facilitate scientific exchange, collaboration and interactions between participants as well as their organisations in advancing science and technology based on rheology.

Athens is reputed as the “cradle of western civilisation” and boasts some of the most important ancient monuments in the world. We hope all international delegates will experience the important attractions that Athens has to offer during their stay in the city.

We are indebted to the members of the Organising and Scientific Committees and other individuals for their commitment and hard work in order to make this Conference a successful event.

Evan Mitsoulis
Conference Chair
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HSR 2004 Organising Committee

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HELENIC SOCIETY OF RHEOLOGY

The Hellenic Society of Rheology (HSR) was officially formed on June 16th, 1996, in Athens, Greece, by the Greek judicial authorities having the necessary core of the first 20 founding members. Thus came into fruition the original attempts by the late Professor Tasos Papanastasiou to organize the Greek Rheological community and the many scientists and engineers who practice rheology in their professional careers.

Rheology enters in some form into almost every study of material properties. Many physicists, chemists, engineers, biologists and mathematicians find a common meeting ground in the Society’s meetings and publications. It is a small society compared to many others, membership currently being about 40. Membership in HSR is open to all researchers in the field, and to all persons who feel the activities of the Society advance their professional development. The membership represents a wide spectrum of individuals from academic, industrial, and governmental institutions whose activities include both phenomenological and molecular theories, instrumentation, the study of many types of materials such as polymers, metals, petroleum products, rubber, paint, printing ink, ceramics and glass, foods, biological materials, floor preparations and cosmetics, and a wide range of practical applications.

The Hellenic Society of Rheology is one of the fifteen (15) founding members of the European Society of Rheology. By virtue of this affiliation, all Members of the Society receive the ESR’s Newsletter. The Society is also a member of the International Committee on Rheology, which organizes the International Congress on Rheology, held every four years.

Please visit the HSR website at:

http://esperia.iesl.forth.gr/~hsr/HSR.html

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**HSR 2004 Program Summary**

**Sunday, June 27, 2004**

17:00-19:00  Registration, Hotel Armonia  
20:30-22:30  Cocktail Party and Buffet by the Hotel Armonia Pool

**Monday, June 28, 2004**

08:30-09:00  Registration  
09:00-09:05  Welcome remarks  
09:05-09:50  Plenary presentation by Prof. Kirk Valanis  
09:50-10:30  2 presentations  
10:30-11:00  Coffee break  
11:00-13:00  6 presentations  
13:00-15:00  Free for lunch  
15:00-16:40  5 presentations  
16:40-17:00  Coffee break  
17:00-18:00  2 presentations + HSR Business Meeting  
18:00-20:30  Break  
20:00-23:00  Banquet

**Tuesday, June 29, 2004**

08:45-09:30  Plenary presentation by Prof. A. Beris  
09:30-10:30  3 presentations  
10:30-11:00  Coffee break  
11:00-13:00  6 presentations  
13:00-15:00  Free for lunch  
15:00-16:40  5 presentations  
16:40-17:00  Coffee break  
17:00-18:00  2 presentations  
18:00  End of Conference

-------------------------------------------------------------------------------------------------------------------

**Total:**

1 Plenary presentation by Prof. K. Valanis (45 min)  
1 Plenary presentation by Prof. A. Beris (45 min)  
31 oral presentations (20 minutes each)  
17 Posters
Monday, June 28, 2004

08:30   Registration
09:00   Welcome Remarks

Morning session I: Rheology and Rheometry

Plenary Talk
09:05-9:50  Kirk Valanis
09:50-10:10  A.D. Gotsis, C. Tsenoglou
"Long chain branches and the rheology of polypropylene"
10:10-10:30  E. Mitsoulis, S.G. Hatzikiriakos
"The effect of slip in the flow of a branched PP melt: simulations and experiments"

10:30-11:00  Coffee break

Morning session II: Rheology and Rheometry

"Quantitative characterisation of human bloods under rheometric flows"
11:20-11:40  G. Gotzamanis, C. Tsitsilianis
"Hydrophobically end-capped cationic polyelectrolytes and their rheological properties in salt-free aqueous solutions"
11:40-12:00  N. Miskolczi, L. Bartha, Gy. Deak, Sz. Biro, A. Geiger, B. Jover
"The change of rheological properties of waste polymers in thermal degradation"
12:00-12:20  A. Guštin, E. Mitsoulis, A. Zupančič
"Polyamide-6: pressure drop estimation through wire mesh filters and spinnerets"
12:20-12:40  W.M. Holmes, P.T. Callaghan, D. Vlassopoulos, J. Roovers
"Observation of shear banding in ultrasoft colloidal gels"
12:40-13:00  G. Petekidis, V. Carrier, D. Vlassopoulos, P.N. Pusey, M. Ballauff
"Rearrangements and yielding in glasses of hard and soft colloids"

13:00-15:00  Free for lunch

Afternoon session I: Structure and Dynamics of Polymers

15:00-15:20  C. Tsitsilianis, F. Bossard, N. Stavrouli, V. Sfika
"The rheology of a physical gel formed by double hydrophilic triblock copolymers"
"Rheology and morphology of associutive telechelic polyelectrolytes"
15:40-16:00  H. Mendil, P. Baroni, L. Noirez
Analysis of shear-induced nematic-isotropic transition in side-chain liquid crystal polymer

16:00-16:20  A. Geiger, Sz. Biro, N. Miskolczi, L. Bartha, Gy. Deak
Changing of viscosity of rubber bitumen in storage time

Performance of asphalt rubber versus shear stress

16:40-17:00  Coffee break

Afternoon session II: Colloids and Suspensions

17:00-17:20  G.E. Yakubov, V. Michailidou, B. Loppinet, G. Fytas, J. Ruhe
Dynamics of end-grafted polymer brushes: an evanescent wave dynamic light scattering study

17:20-17:40  T.B. Goudoulas, E.G. Kastrinakis, S.G. Nychas
Characteristic viscosities and a stress model fitting on consecutive flow curves of dense lignite-water suspensions

17:40-18:00  HSR Business Meeting

18:00  End

20:00-23:00  Banquet
Tuesday, June 29, 2004

Morning Session I: Numerical Simulations

**Plenary Talk**

08:45-9:30  A.N. Beris
*Recent advances in DNS of turbulent viscoelastic channel flows: Understanding polymer-induced drag reduction*

09:30-09:50  K.D. Housiadas, A.N. Beris
*An efficient spectral technique for direct numerical simulations of viscoelastic turbulent channel flow*

09:50-10:10  K. Foteinopoulou, V.G. Mavrantzas, J. Tsamopoulos
*Numerical simulation of bubble growth during filament stretching of Newtonian and viscoelastic fluids*

10:10-10:30  Y. Dimakopoulos, J. Tsamopoulos
*Transient displacement of viscoelastic liquid by air*

10:30-11:00  Coffee break

Morning Session II: Numerical Simulations

11:00-11:20  L. Pamela Cook, R.J. Braun
*Thin films and viscoelasticity*

11:20-11:40  G. Karapetsas, N. Chatzidaki, M. Pavlidis, J. Tsamopoulos
*Transient squeeze flow of viscoplastic liquids*

11:40-12:00  M. Chatzimina, G.C. Georgiou, E. Mitsoulis, R.R. Huilgol
*Time-dependent Couette and Poiseuille cessation flows of Bingham fluids*

12:00-12:20  A.N. Alexandrou, G.C. Florides, G.C. Georgiou
*Simulations of Herschel-Bulkley flow in compression molding*

12:20-12:40  E. Mitsoulis, S. Sofou
*Simulations of Herschel-Bulkley flow in roll coating*

12:40-13:00  J. Vlachopoulos, D. Strutt
*Modeling challenges in plasticating screw extrusion*

13:00-15:00  Free for lunch

Afternoon Session I: Molecular Modeling

15:00-15:20  I.A. Bitsanis, A.N. Rissanou, D. Vlassopoulos
*The origin of thermally-induced vitrification in suspensions of multi-arm star polymers: a molecular dynamics study*

15:20-15:40  N.Ch. Karayiannis, V.G. Mavrantzas
Branch point friction and its role in the dynamics of H-shaped polyethylene melts as probed by long molecular dynamics simulations

15:40-16:00  **G. Tsolou, V.G. Mavrantzas**  
*Atomistic simulation of the dynamics of cis-1,4 polybutadiene and its dependence on temperature and pressure*

16:00-16:20  **A. Tzavaras**  
*Structural properties of hyperbolic relaxation, an example from viscoelasticity with memory to polyconvex elastodynamics*

16:20-16:40  **H. Pleiner, M. Liu, H.R. Brand**  
*A Physicist’s view on constitutive equations*

**Coffee break**

**Afternoon Session II: Experimental, theoretical and computational fluid dynamics**

17:00-17:20  **K. Tsiglifis, N. Pelekasis**  
*Weak viscous oscillations and collapse of laser and acoustic bubbles*

17:20-17:40  **A.N. Spyropoulos, A.G. Boudouvis**  
*Finite element computations with parallel Newton/Krylov solvers of Beowulf clusters*

17:40-18:00  
18:00  **End**
Poster Session:

1. C.V. Boas, C. Tzoganakis
   Rheological properties of controlled-rheology metallocene polypropylenes

2. L. Slemenik Perše, M. Žumer, A. Zupančič, A. Bouras, A. Boudouvis, E. Mitsoulis
   Laminar mixing of non-Newtonian fluids in agitated vessels with baffled impellers: experiments and simulations

3. E. Mitsoulis, C. Papoulias
   The effect of fiber-glass content on the rheology of semi-concentrated polymer composites

4. E. Mitsoulis, P. Kotsos
   Rheological effects in wire coating

5. E. Mitsoulis, A. Matsoukas
   Squeeze flow of yield-stress fluids: theory and experiments

6. E. Mitsoulis, S. Galazoulas
   The effect of rheology on the drag and stability of cylinders moving in yield-stress fluids

7. E. Mitsoulis, A. Psarreas, G. Ntagas
   Start-up Couette and Poiseuille flows of yield-stress fluids

8. Y.A. Altukhov, V.N. Pokrovskii
   Numerical simulation of polymeric fluid flows

9. M.A. Makarova, G.V. Pyshnogray
   The mesoscopic theory of linear polymer solutions and melts dynamics

    Depletion attraction in mixtures of soft colloids and linear polymers in solution

11. M. Dressler, B.J. Edwards
    Rheology of polymer blends with a narrow droplet size distribution

12. C. Clasen, W.-M. Kulicke
    Determination of structure-property-relationship of polymers in solution

13. M. Kapnistos, D. Vlassopoulos, L.G. Leal, J. Roovers
    Rheology of model branched polymers

14. P.K. Rozakeas
    Electrokinetically enhanced flow of coal-water suspensions in a pipe with a helical anode-cathode design embedded in the pipe wall

15. E. Taliadorou, E. Stiakakis, G. Georgiou, D. Vlassopoulos
    A structural model for thixotropic fluids exhibiting yield stress

16. I. Katsampas, C. Tsitsilianis, D. Vlassopoulos
    Stimuli responsive physical gel formed by ABC telechelic polyelectrolyte

17. B. Abu-Jdayil, H.A. Mohameed
    Effect of Dead Sea salt on the rheological behavior of hair shampoo
ABSTRACTS

PLENARY LECTURES
Kirk Valanis
Endochronics / University of Portland
Portland, Oregon, USA

Notes:
Recent advances in DNS of turbulent viscoelastic channel flows: Understanding polymer-induced drag reduction

A.N. Beris
Department of Chemical Engineering
University of Delaware, USA

The recent progress in understanding polymer-induced drag reduction in turbulent flows through Direct Numerical Simulations (DNS) is going to be presented. High performance parallel computations have generated results for various turbulence and conformation statistics for the turbulent channel flow of a dilute polymer solution modeled from first principles with the FENE-P, Giesekus and Oldroyd-B differential constitutive equations.

In particular, our most recent data have helped us elucidate the dependence of drag reduction on the Weissenberg number, polymer concentration, extensibility parameter, the Reynolds number and the polymer model: Drag reduction sets in at a critical value of the Weissenberg number (typically about 8) and continues increasing, albeit at a lower rate, even up to the highest value examined (125). Drag reduction values as high 50% are predicted. The drag reduction increases as the Weissenberg number, molecular extensibility and the polymer concentration increase. In addition, the drag reduction is significantly enhanced if a non-zero second normal stress is predicted by the model in simple shear flows, such as happens with the Giesekus model. In contrast, it was found that for constant rheological parameters (non-dimensionalized with respect to turbulent units) the drag reduction remains practically unchanged as the friction Reynolds number increases from 125 to 590. Nevertheless, the details of the near the wall turbulent structure change continuously as the friction Reynolds number increases up to 395.

The main effect of viscoelasticity is shown to be the strengthening of the largest size turbulent structures which become much more coherent with a dynamics that changes at an appreciably lower rate than for the equivalent Newtonian structures. Our parametric study strongly suggests that this feature develops due to an enhanced resistance to extensional deformation induced due to viscoelasticity and it results to a lower energy transfer from the wall to the turbulent core, thus explaining the drag reduction. The numerical results provide thus evidence and in depth analysis to a mechanism proposed first in the 60s by Lumley and Metzner based on experimental observations.

Notes:
COLLOIDS AND SUSPENSIONS
Electrokinetically Enhanced Flow of Coal-Water Suspensions in a Pipe with a Helical Anode-Cathode Design Embedded in the Pipe Wall

P.K. Rozakeas

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It has been experimentally demonstrated that the application of electrokinetic techniques in the continuous flow of coal-water suspensions in various anode-cathode pipe geometries effectively causes a significant reduction in the wall shear stress. This reduction in the frictional force at the cathodic pipe wall surface is attributed to a decrease in the concentration of coal particles in this flow region caused by the migration of negatively charged coal particles towards the anodic surface(s). It is possible to reduce the specific pumping energy requirements of coal-water suspensions flowing in pipes by as much as an order of magnitude.

The disadvantage of flow-intrusive geometries is the reduction in effective flow area as well as an increase in frictional surface area caused by the placement of the anode(s) within the flow region. The results of the preliminary experimental work carried out on a non-intrusive helical anode-cathode pipe design is presented and compared with data generated from various concentric and eccentric annular flow geometries.

Fine coal particles with a mean diameter of 17.7 micron were produced by milling a low rank bituminous black coal which was followed by a sieving process that eliminated coal particles that were greater than 75 micron in diameter.

Notes:
Aspects of dense Lignite-Water Slurry (LWS) rheology were investigated using a controlled stress rheometer, with a parallel plate geometry. The achieved solids volume fraction was up to 0.425. The particle size distribution was polydispersed having sizes in the range of 1 \( \mu \text{m} \) to 300 \( \mu \text{m} \). The investigation covered a wide range of shear stresses (up to 120 Pa) and shear rates (from \( 10^{-3} \text{ s}^{-1} \) to \( 10^{2} \text{ s}^{-1} \)) while the flow curves were carried out for various ascending times. Due to consecutive flow loops, in certain cases, the total shearing time was up to 70 min. Characteristic viscosities, during the flow curves, were defined and studied with respect to the measurement procedure. It was found that non-dimensional ratios of these viscosities were correlated with characteristic time ratios of the flow loop in an exponential way. For the lower applied shear stress and for all flow loop times, a thixotropic behavior was evident. However, for the higher applied shear stress the suspension showed an antithixotropic behavior in the low shear region (\( 60 \text{ s}^{-1} \)). In the ascending part of the flow curves a transition point was observed, which designates a change in flow curve slope. This observation is in accordance with the results of the recent study by Cheng (2003) in thixotropic fluids. Furthermore, due to the yield stress appearance in LWS, by following the Cheng (2003) approach, a combination of viscosity and stress models seems to fit the experimental data well enough in the transition part as well as in the low shear region (\( \gamma < 1 \text{ s}^{-1} \)). This rheological response is attributed to different structure formation according to the shear conditions in addition to the particle properties, i.e. irregular shape and particle size distribution.

Notes:
In Jordan, a growing industry has been established to produce different types of Dead Sea (DS) cosmetics that have Dead Sea salt (contains mainly $\text{NaCl}, \text{KCl}, \text{MgCl}_2$) in their formulas. In this work, the effect of DS salt on the rheology of hair shampoo containing the sodium lauryl ether sulfate as a main active matter was studied. The effects of DS salt and active matter concentration, and the temperature and time of salt mixing on the rheological properties of hair shampoo were investigated. The salt free shampoo showed a Newtonian behavior at “low active matter” (LAM) and shear thinning at “high active matter” (HAM). The presence of DS salt changed the rheological behavior of LAM shampoo from Newtonian (for the salt free shampoo) to shear thinning. On the other hand, at the behavior of HAM shampoo switched from shear thinning to Newtonian behavior in the presence of high concentration of DS salt. The addition of DS salt increased the apparent viscosity of shampoo to reach a maximum value that corresponded to a salt concentration of 1.5 wt%. Further addition of DS salt led to a decrease in the shampoo viscosity to reach a value less than that of the salt free sample at high salt concentration. The mixing process increased the apparent viscosity of salt free shampoo. In addition, the rheological properties were tested during a 28-day storage period at three different storage temperatures (cold, room temperature and accelerated conditions). The apparent viscosity of salt free shampoo decreased with storage time, while the rheological behavior changed from shear thinning to Newtonian. In the presence of 1.5 wt% DS salt, the rheological behavior of shampoo samples did not change over the 28-day storage period.

Notes:

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Depletion Attraction in Mixtures of Soft Colloids and Linear Polymers in Solution

E. Stiakakis¹, G. Petekidis¹ D. Vlassopoulos¹, C. N. Likos², J. Roovers³, H. Iatrou⁴, N. Hadjichristidis⁴

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³NRC, Institute for Chemical Process and Environmental Technology, Ottawa, Ontario, Canada
⁴Department of Chemistry, University of Athens, Athens, Greece

The dynamics in solutions of multi-arm stars (with 128 arms) are studied in the presence of linear homopolymer chains of the same kind (polybutadiene) by dynamic light scattering. The former can be described well as a soft spherical particle with repulsive inter-particle potential while the latter act as a depletant of the soft spheres. The depletion interaction induced by the presence of the linear polymer chains leads to the formation of clusters of the multi-arm stars in dilute solutions of the latter and close to the overlap concentration of the former. This finding directly suggests that due to the depletion interaction the inter-particle potential between two stars should have an attractive part as suggested by theory. Moreover, at high star volume fractions, where in the absence of linear polymer chains a physical gel has been observed in good solvents, the addition of small amounts of linear chains leads to a liquidification of the gel. Such phenomena, at low and high particle volume fractions reveal the colloidal character of the multi-arm star polymer and resemble both the well known depletion induced phase separation in colloid-polymer mixtures as well recent reentrant phenomena observed in colloidal systems in the glass regime.

Notes:
EXPERIMENTAL, THEORETICAL, AND COMPUTATIONAL FLUID DYNAMICS
The fashion by which a bubble oscillates and collapses plays an important role in ultrasound based applications and in phenomena like sono-luminescence. In the present study, a hybrid boundary-finite element method is used in order to follow the shape deformation and collapse of axisymmetric bubbles in the presence of weak viscous effects, in response to an initial elongation and/or pressure disturbance. The description of the bubble’s interface is based on a Lagrangian representation. B-cubic splines are used for the discretization of the unknown functions along with the 4th order Runge-Kutta method for the time integration. The mesh and time step are updated during the simulation in order to capture curvature variations. The effect of small viscosity is included in the computations by retaining first-order viscous terms in the normal stress boundary condition and satisfying the tangential stress balance. Construction of the system matrix is parallelized for optimal code efficiency. In many situations, the simulation with weak viscous effects reveals a different bubble behavior from that based on the inviscid model. This has to do with the fact that viscosity eliminates the higher modes that tend to be excited in the inviscid model as time progresses. Several different modes of bubble collapse are obtained. Preliminary results show that an initially elongated axisymmetric bubble with much larger internal pressure than the static pressure, tends to collapse in the direction of initial elongation. These results conform well with recent experiments with laser bubbles.

Figure 1. Deformation & break-up of an inviscid laser bubble; R, Ebubl, Pinf, parameters measuring initial elongation, internal pressure disturbance and bubble size, for an air bubble in water at atmospheric pressure.

Notes:
In the last decade parallel processing on Beowulf clusters became a popular alternative way for large scale computations due to their low cost compared with the cost of parallel supercomputers. The most important factor that limits the parallel efficiency of an algorithm running on a cluster is the low bandwidth and high latency of the network that interconnects the computers. Specially designed parallel algorithms must be applied that have low communication overhead. A parallel inexact Newton method for Galerkin/finite element computations on clusters is presented. This method is based on a parallel preconditioned Krylov-type iterative solver for the solution of large, sparse and non-symmetric equation systems and a preconditioning method for the inexact Newton steps. The choice of the two preconditioners and their parallel implementation targets on the scalability of the parallel Newton/Krylov algorithm when it is applied on a cluster. Two important aspects of the method are addressed: the storage of the coefficient matrix of the system and of the preconditioning matrix, and the performance of the preconditioners. The matrix storage affects the parallel efficiency of the matrix vector product. The preconditioning techniques contribute to the parallel efficiency and are of critical importance for the convergence rate of the Newton/Krylov solver. The performance of the method is analyzed in terms of parallel speedup, storage efficiency and convergence rate.

Notes:
Laminar Mixing of Non-Newtonian Fluids in Agitated Vessels with Baffled Impellers: Experiments and Simulations

L. Slemenik Perše¹, M. Žumer¹, A. Zupančič¹, A. Bouras²a, A. Boudouvis²a, E. Mitsoulis²b

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Agitated vessels are primarily used as mixing devices for several non-Newtonian fluids with the aim to improve the mixing properties. Close-clearance impellers are a convenient means of achieving this. In the experimental part of this work, the mixing vessel was equipped with a radial baffled impeller of vane geometry. The vane geometry offers effective rheological measurements for the fluids that would otherwise display large slip effects at smooth walls. In mixing vessel vane geometry was chosen in order to measure the viscosity function of non-Newtonian fluids during mixing process. The materials were Newtonian motor-oil fluids for reference, and non-Newtonian pseudoplastic, viscoplastic, and viscoelastic fluids, such as polymer carboxymethyl cellulose (CMC) solutions and polysaccharide welan gels. Rheological characterization has produced their corresponding viscosity results and other related properties.

The mixing experiments have shown that in the laminar flow regime, a unique power curve was obtained when the Newtonian results were plotted vs. Reynolds number. They followed the well-known relation of $Re^{-1}$. For non-Newtonian fluids, shear constant $K_s$ was calculated from experimental data. The viscosity functions, obtained from mixing experiments corresponded to that measured by rheometer. Numerical simulations, using the corresponding non-Newtonian modes, are then employed to understand this important difference in behaviour. Viscoplastic results show the presence of a viscoplastic cavern of no deformation, and its size is related to the fluid properties and in particular the yield stress.

Notes:
MOLECULAR MODELING
The Origin of Thermally-Induced Vitrification in Suspensions of Multi-Arm Star Polymers: A Molecular Dynamics Study

I.A. Bitsanis\textsuperscript{1}, A.N. Rissanou\textsuperscript{1,2}, D. Vlassopoulos\textsuperscript{1,3}

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\textsuperscript{3}Dept. of Materials Sci. & Eng., Univ. of Crete, Heraklion, Crete, Greece

Dense suspensions of multi-arm star polymers are known to develop liquid-like microstructure, which has been attributed to the similarities between high functionality stars and colloidal particles interacting via soft, long ranged potentials. Experimental studies\textsuperscript{*} reported a counter-intuitive solidification of suspensions with star functionality \( f = 128 \), upon increase of the temperature in marginal solvents. We present our results from Molecular Dynamics (MD) simulations of dense suspensions of multi-arm star polymers. Star polymers are modeled as ‘soft-spheres’ interacting via a theoretically developed potential of mean field. Our results show a transition towards a ‘glassy’ state at a temperature very close to the one reported experimentally. The features of the transition are consistent with those of \textit{ideal} glass transitions, as described by \textit{ideal} Mode Coupling Theory. Furthermore, our findings illustrate the road to vitrification for these soft-colloidal suspensions. Higher temperatures result in arm expansion that causes jamming and more than compensates for faster short time, temperature induced kinetics.


Notes:
We report results from the first atomistic molecular dynamics (MD) simulation of the simplest long-chain branched polymer, the H-shaped polyethylene. Initial configurations for the simulations were provided by a novel MC algorithm, based on the double-bridging and intramolecular double rebridging moves. The simulations have been executed in the NPT ensemble for times longer than 2ns and covered a wide variety of chain systems, crossing also above the entanglement molecular weight, $M_e$. By monitoring the mean-square displacement (msd) of the center of mass and of the branch points as a function of time, our simulations have provided information about the self-diffusion behavior of the H-shaped chains. In the long-time regime where Fickian diffusion is reached, the data have shown that the center-of-mass diffusivity follows faithfully that of the branch points: For systems below $M_e$, the two msd curves travel one next to the other in time (i.e., in parallel) right from the beginning. For systems above $M_e$, this happens after an initial sub-diffusive time regime is elapsed. It is the first time that such a result is documented, validating from first principles one of the most important assumptions of the Larson-McLeish pompom theory that all friction in an H-shaped polymer is concentrated at the branch points.
Atomistic simulation of the dynamics of cis-1,4 polybutadiene and its dependence on temperature and pressure

Georgia Tsolou\textsuperscript{1,2} and Vlasis G. Mavrantzas\textsuperscript{1,2,3}

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Results are presented for the dynamic properties of cis-1,4 polybutadiene (cis-1,4 PB) systems obtained through detailed atomistic molecular dynamics (MD) simulations in the NPT statistical ensemble. The MD simulations are carried out with the parallel LAMMPS software \cite{1} using as initial configurations thoroughly equilibrated model cis-1,4 PB structures obtained with the end-bridging Monte Carlo algorithm \cite{2}. Systems ranging in molecular length from C\textsubscript{32} to C\textsubscript{400} have been simulated at $T=413$ K (i.e., above the melting point) and $P=1$ atm. Our MD simulation data show that, above C\textsubscript{200}, the chain self-diffusion coefficient $D$ of the simulated cis-1,4 PB melts exhibits a clear change in its power-law dependence on the molecular weight ($M$), significantly deviating from a Rouse (where $D \sim M^{-1}$) towards a reptation-like (where $D \sim M^{-2.1}$) behavior. Results are also presented about the temperature and pressure dependence of the dynamic properties of cis-1,4 PB systems; they have been obtained by subjecting strictly monodisperse cis-1,4 PB samples to MD simulations under different isothermal and isobaric conditions. The relative contributions of density and thermal energy to the local and global relaxation of cis-1,4 PB will be presented and discussed in detail.

References
\cite{1} [www.sandia.gov]

Notes:
Structural properties of hyperbolic relaxation, an example from viscoelasticity with memory to polyconvex elastodynamics

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The subject of this talk is the approximation of the equations of polyconvex elastodynamics via models of viscoelasticity with memory. The process can be embedded to the framework of materials with internal variables, where the internal variable is the viscoelastic stress. An estimate analogous to the H-theorem is developed for this model and a globally defined entropy is constructed for the relaxation process under a subcharacteristic condition. Moreover, we develop a "strong dissipation" estimate which shows that the approximation has the same efficacy as the usual viscosity approximation. A third relative entropy estimate is constructed which shows stability of the approximation procedure for classical solutions type. The last step requires the use of embedding the equations of polyconvex elastodynamics to an extended symmetric hyperbolic system. (joint work with C. Lattanzio, Univ. de l'Aquila).

Notes:
A Physicists’ View on Constitutive Equations

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Hydrodynamic equations for various kinds of complex fluids have been derived rigorously using general physical laws and principles. This hydrodynamic method is generalized to include slowly relaxing quantities, in particular those describing viscoelasticity. We start with the nonlinear hydrodynamic equations for elastic media derived from basic physical principles. For the Eulerian strain tensor the lower convected time derivative is obtained, unambiguously. Adding a relaxation term the permanent elasticity is transformed into viscoelasticity [1,2], where both, the short time and the long time limit, are given correctly. The dynamic equation for the strain tensor obtained that way still shows the lower convected derivative universally. It covers the usual non-Newtonian effects, like shear thinning, strain hardening, stress overshoot, normal stress differences and Weissenberg effect, non exponential stress relaxation, etc. When brought into the more familiar form of a dynamic equation for the stress tensor ("constitutive equation"), it comprises most of the well-known ad-hoc models (Maxwell, Oldroyd, Johnson-Segalman) and is even more general in structure than those. Nevertheless, it imposes also some restrictions on, and reveals some interdependencies of, the various non-Newtonian contributions that are otherwise introduced heuristically. It is shown how these contributions originate from (nonlinear) elasticity, viscosity, strain relaxation and convection. We also discuss [3] the connection to those descriptions of viscoelasticity that utilize an orientational order parameter.

Rheology of polymer blends with a narrow droplet size distribution

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A Hamiltonian framework of non-equilibrium thermodynamics is adopted to construct a set of dynamical continuum equations for a polymer blend with a narrow droplet size distribution that is assumed to obey a Weibull distribution function. The variable droplet distribution is described in terms of two thermodynamic variables: the droplet shape tensor and the number density of representative droplets. A Hamiltonian functional in terms of the thermodynamic variables is introduced and a set of time evolution equations for the system variables is derived. Sample calculations for a blend with variable droplet distribution are performed and the effect of flow on the rheology, droplet morphology, and on the droplet distribution are discussed. It is found that deformation increases the dispersity of the droplet morphology for all flows investigated herein.

Notes:
(3-P) Poster Session

The Mesoscopic Theory of Linear Polymer Solutions and Melts Dynamics

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The work begins with an introduction to the dynamics of single macromolecule in the entangled system. Diffusive and anisotropic expressions for the drag forces mechanisms of relaxation of macromolecules are considered. Rheological equation of the state is obtained from microstructural representations about dynamics of macromolecule. Initial value of the shear viscosity; initial value of relaxation time; two factors of anisotropy, which in their turn depend on concentration of polymer and its molecular weight are the parameters of this rheological model. At superposition of small oscillations on fixed shift flow in direction, parallel a shift, the behaviour of polymeric medium is characterized by the module of a shift and module of losses, which are functions of frequency of oscillations and velocity of a shift. Numerical calculations have allowed to establish presence of critical values of a velocity of a shift, at which excess the module of a shift in the field of small frequencies becomes negative, that was observed in experiments earlier. The method of perturbations of the small parameter determining the anisotropy of the properties of linear polymers is used to determine the velocity profile and rate for steady flow in a round tube. It is shown that for the four-parameter rheological model considered, the stress state of the Poiseuille flow along with the tangential shear stress is characterized by the first and second differences of normal stresses.

Notes:
NUMERICAL SIMULATIONS
Recent advances in DNS of turbulent viscoelastic channel flows: Understanding polymer-induced drag reduction

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The recent progress in understanding polymer-induced drag reduction in turbulent flows through Direct Numerical Simulations (DNS) is going to be presented. High performance parallel computations have generated results for various turbulence and conformation statistics for the turbulent channel flow of a dilute polymer solution modeled from first principles with the FENE-P, Giesekus and Oldroyd-B differential constitutive equations.

In particular, our most recent data have helped us elucidate the dependence of drag reduction on the Weissenberg number, polymer concentration, extensibility parameter, the Reynolds number and the polymer model: Drag reduction sets in at a critical value of the Weissenberg number (typically about 8) and continues increasing, albeit at a lower rate, even up to the highest value examined (125). Drag reduction values as high 50% are predicted. The drag reduction increases as the Weissenberg number, molecular extensibility and the polymer concentration increase. In addition, the drag reduction is significantly enhanced if a non-zero second normal stress is predicted by the model in simple shear flows, such as happens with the Giesekus model. In contrast, it was found that for constant rheological parameters (non-dimensionalized with respect to turbulent units) the drag reduction remains practically unchanged as the friction Reynolds number increases from 125 to 590. Nevertheless, the details of the near the wall turbulent structure change continuously as the friction Reynolds number increases up to 395.

The main effect of viscoelasticity is shown to be the strengthening of the largest size turbulent structures which become much more coherent with a dynamics that changes at an appreciably lower rate than for the equivalent Newtonian structures. Our parametric study strongly suggests that this feature develops due to an enhanced resistance to extensional deformation induced due to viscoelasticity and it results to a lower energy transfer from the wall to the turbulent core, thus explaining the drag reduction. The numerical results provide thus evidence and in depth analysis to a mechanism proposed first in the 60s by Lumley and Metzner based on experimental observations.

Notes:
Simulations of time-dependent fully dimensional turbulent viscoelastic flows have been routinely performed during the last few years for friction Reynolds numbers up to 220. However, there is a big need to perform the simulations for higher friction Reynolds number in order to validate the results against experimental data. As the Reynolds number increases the computational requirements are also increase. Moreover, numerical instabilities often develop making the simulations either unfeasible or much more computationally expensive to perform. Therefore, the efficiency and stability of the numerical algorithm being used is of great importance.

We have developed here a significantly improved numerical algorithm, based on a spectral approximation as was successfully used before. Briefly, the features of the new algorithm are: (a) full spectral approximations for all the dependent variables, (b) totally implicit time integration scheme (of variable accuracy; from 1st up to 6th order), (c) dealiasing of the calculations in both the homogeneous and non-homogeneous directions, (d) machine satisfaction of the divergence-free velocity condition, (e) exact enforcement of the total force balances in all directions.

The new algorithm has enabled us to extend the simulations for much wider range of viscoelasticity parameter values as well as for different viscoelastic models beyond FENE-P like Giesekus and Oldroyd-B. Among others things, we will present the influence of: (a) the order of accuracy in the time integration (b) the dealiasing (c) the total integration time (d) of the enforcement of the total force balances and the machine accuracy satisfaction of the continuity equation on the stability and accuracy of the calculations.
Numerical calculation of bubble growth during filament stretching of Newtonian and viscoelastic fluids

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We present results concerning bubble deformation in Newtonian and viscoelastic filaments undergoing stretching. In the application discussed here, the deformation of a single (initially spherical) bubble is considered inside a filament (having initially the shape of a cylinder with uniform radius) whose upper plate is pulled along its axis with a constant velocity. The governing equations consist of the momentum, continuity and constitutive equations and the free surface boundary conditions at the bubble-liquid and liquid-air interfaces. These are solved by a finite element/Galerkin method coupled with a 1st order implicit Euler scheme for time integration. Both a Newtonian and a viscoelastic medium modeled as a Phan-Thien/Tanner liquid are addressed. In the latter case, the EVSS-G technique is used to separate elastic and viscous contributions to the stress tensor together with an SUPG discretization of the constitutive equation. Our numerical calculations provide information on the dependence of bubble and filament deformation on the capillary and Deborah numbers, relative bubble size, and proximity of the bubble to and liquid slippage on the substrate. Efforts are currently in progress to address the deformation of several bubbles in the drawn filament, through the implementation of the robust elliptic grid generation scheme.

Notes:
Transient Displacement of Viscoelastic Liquids by Air

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We examine the transient displacement by air of viscoelastic liquids, as it arises in the Gas-Assisted Injection Molding (GAIM) Process. The liquid occupies axisymmetric tubes of constant or variable cross section, either completely (when one moving interface arises) or partially (when two moving interfaces and a moving contact line arise, necessitating the use of a proper slip condition). For the simulations we combine our recently advanced quasi-elliptic grid generation scheme for computing the highly deforming liquid boundaries, the mixed finite element technique and the Discontinuous Galerkin method for computing the viscoelastic stresses. In our parametric study we examine the effects of the elastic and inertia forces and the solvent to polymer viscosity ratio. Results using the PTT and Giesekus constitutive models show that the thickness of the remaining film increases as the Deborah number increases and remaining fluid fractions greater than 0.60 (the Newtonian limit) arise, in agreement with experiments. Increasing the viscosity ratio decreases the effects of elasticity. Similar is the effect of the Reynolds number which may also result in variable film thickness and a tip splitting instability.

Notes:
Problem formulation, scaling, analysis and computation of a single layer thin film of a viscoelastic solution is presented. The problems discussed arise from physiological applications. The models describing these free boundary thin films are examined under a variety of boundary conditions including moving boundaries. In this context several models for the viscoelastic fluid are considered, which predict shear-thinning and elastic effects, and embedded within a Newtonian solvent. The lubrication approximation is used. Capillarity at the free surface and as well gravitational effects are considered. Asymptotic and computational results will be presented describing the stability properties of the thin film and the film break-up time. We believe that the results are important for understanding the behavior of the tear film in both normal and "dry" eyes.
Transient Squeeze Flow of Viscoplastic Liquids

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We examine the axisymmetric squeeze-flow of viscoplastic materials placed between two parallel disks using the continuous constitutive model suggested by Papanastasiou and the discontinuous Bingham model. This is the first transient simulation of such liquids and allows us to also determine the shape of the liquid/air interface. When the interface is initially located inside the disks we follow its displacement towards their edge. We have also carried out computations when the liquids exceed the boundaries of the discs. We employ the mixed finite element method coupled with a quasi-elliptic mesh generation scheme in order to follow these large deformations. The material yields in part of the domain as the disks approach each other, departing from the corresponding Newtonian solution. Unyielded material arises around the two stagnation points of flow at the disk centers verifying previous steady state calculations. The unyielded region increases with the Bingham number, but decreases with time. If wall-slip is present, the unyielded region decreases significantly and may even totally disappear.

Notes:
Time-Dependent Couette and Poiseuille Cessation Flows of Bingham Fluids

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We solve the cessation Couette and Poiseuille flows of a Bingham fluid using the regularized constitutive equation proposed by Papanastasiou. Our objective is to predict the finite stopping times and make comparisons with theoretical upper bounds provided in the literature. The calculations confirm that the stopping times are indeed finite when the yield stress is nonzero. The decay of the volumetric flow rate, which is exponential in the Newtonian case, is accelerated and eventually becomes linear as the yield stress is increased. Preliminary comparisons of the calculated stopping times of plane Poiseuille flow with the upper bound estimate of Huilgol et al. are provided.

Notes:
Simulations of Herschel-Bulkley Flow in Compression Molding

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The typical compression molding flow experiment of a semisolid material is simulated using the regularized Herschel-Bulkley constitutive equation and a mixed-Galerkin finite-element method. The objective is to contribute to the development of a methodology, which relates the simulations with the experimental shapes of the test specimen, in order to determine the material constants of semisolid slurries. The time-dependent flow is solved either under constant load or under constant velocity, and the effects of the material and flow parameters on the evolution of the specimen shape and on the yielded and unyielded regions are investigated. A special damage model, which takes into consideration the physical behaviour of the semisolid materials, has also been developed and integrated in the simulation code.

Notes:
Roll coating is a process used in many industries, such as the paper, plastics, rubber, and steel industries, for the production of thin layers of specific thickness and final appearance, coated over a moving web. The rheology of many materials used in roll coating is non-Newtonian. These materials may exhibit either pseudoplastic (shear-thinning or -thickening) or viscoplastic (presence of a yield stress) rheological behaviour.

In the present work, the Lubrication Approximation Theory (LAT) is used to provide numerical results in roll coating over a moving flat web. The Herschel-Bulkley model of viscoplasticity is used, which reduces with appropriate modifications to the Bingham, power-law and Newtonian models. Results are obtained for such quantities as coating thickness, split location, pressure distribution, stresses, forces, torque, power input to the roll and adiabatic temperature rise between the coating roll and the coated web. A full parametric study is undertaken for the dimensionless power-law index (in the case of pseudoplasticity) and the dimensionless yield stress (in the case of viscoplasticity). Both pseudoplasticity and viscoplasticity lead to excess coating thickness over the gap at the nip. All engineering quantities, given in a dimensionless form, increase substantially with the departure from the Newtonian values. For the viscoplastic materials, the yielded/unyielded regions are also shown.

Notes:
Modeling Challenges in Plasticating Screw Extrusion

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Plasticating screw extrusion involves the transport of solid pellets, melting and melt pumping. Although the invention of screw pumping of liquids can be traced back to Archimedes and before, understanding of screw extrusion of polymers is about 50 years old. Beyond the early developments during the 1950’s and the 1960’s there has not been much groundbreaking success in modeling. There are challenges in modeling the forward transport of the compacted solids pellets and melting of them. Extruder designs involving grooved feed sections and barrier screws are even more difficult to describe mathematically. In the presentation, several sets of experimental results will be shown and some modeling assumptions will be described for prediction of flow rates, pressure and temperature build up and solid bed profiles. The long term questions of describing solids transport mainly due to friction and melting under the influence of shear will be discussed.

Notes:
(11-2-P) Poster Session

The Effect of Rheology on the Drag and Stability of Cylinders Moving in Yield-Stress Fluids

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Objects set in yield-stress fluids may or may not move depending on the balance between yield and buoyancy forces. Yield-stress fluids are non-Newtonian, exhibiting both pseudoplastic (shear–thinning or –thickening) and viscoplastic (presence of a yield stress) behaviour. In particular, Carbopol gels are found to obey the Herschel-Bulkley model of viscoplasticity. Some of the objects of a previous experimental study [1] are disks and cylinders moving vertically at very low velocities in a cylinder filled with a Carbopol gel. The experimental results give a stability criterion as a dimensionless yield stress at which there cease to be any movement of the object. Numerical simulations with the continuous viscoplastic model of Herschel-Bulkley/Papanastasiou [2] show the extent and shape of the yielded/unyielded regions in the case of three objects. This type of information is very difficult to extract experimentally. The simulations also provide the drag force exerted on the objects through integration of the pressure and stresses on the object. The results from the simulations are compared with the experiments, and the stability criteria are assessed through a detailed analysis in the range of vanishing velocities.


Notes:
Squeeze Flow of Yield-Stress Fluids:
Theory and Experiments

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The problem of squeeze flow between two circular disks gives rise to the plastometer, which is used in rheometry of non-Newtonian fluids to determine the rheological constants of yield-stress fluids. Such fluids (Carbopol gels, toothpaste, TiO$_2$ particles in treacle) have been used in an experimental setup working under constant-force conditions.

The theory based on Newtonian fluids is inadequate for predictions on yield-stress fluids, as shown in the present experiments. Modifications of the theory based on viscoplastic models are necessary to derive equations able to predict the time-dependent movement of the disks in the plastometer. Numerical simulations have been undertaken based on the Finite Element Method (FEM) for the equivalent problem of constant disk speed. The simulations with the Herschel-Bulkley model show the existence of small unyielded regions only in the center and adjacent to the disks.

Previous equations based on a theory by Covey and Stanmore show the adequacy of such a model for the squeeze force as a function of a dimensionless Bingham number. New results are derived for the case of squeeze flow of yield stress fluids based on the constant-speed problem.

Notes:
(11-4-P) Poster Session

\textbf{Start-Up Couette and Poiseuille Flows of Yield-Stress Fluids}

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Time-dependent flows of yield-stress fluids are of interest in injection molding of composite materials, where various shapes are produced. Aluminum powders with polymer binders are a good example of such materials, since they are known to exhibit a measurable yield stress.

First, the numerical solutions for Newtonian fluids are tested against the well-known analytical solutions in simple Couette and Poiseuille (both planar and axisymmetric) start-up flows. Then the same problems are solved for the Bingham and Herschel-Bulkley models regularized according to Papanastasiou’s modification. The emphasis is on finding the yielded/unyielded lines for different values of the dimensionless Bingham number $Bn$ and the power-law index $n$. The “infinite” times for reaching the steady-state values are also calculated. It is found that the most significant changes occur for shear-thickening yield-stress fluids ($n>1$), which take the most time to fully develop, and the least changes occur for shear-thinning yield-stress fluids ($n<1$), which take the least time to fully develop.

These benchmark results are a prelude for solving more elaborate problems in 2-D and 3-D geometries.

Notes:
Wire coating is a process used in the plastics industry for the production of thin protective layers of specific thickness and final appearance coated over a moving wire. The rheology of many materials used in wire coating is non-Newtonian. These materials may exhibit either pseudoplastic (shear-thinning or -thickening) or viscoplastic (presence of a yield stress) rheological behaviour.

In the present work, both a 1-D analysis based on the Lubrication Approximation Theory (LAT) and a 2-D analysis based on the Finite Element Method (FEM) are used to provide numerical results in an industrial-design wire-coating die. The Herschel-Bulkley model of viscoplasticity is used, which reduces with appropriate modifications to the Bingham, power-law and Newtonian models. Results are obtained for such quantities as pressure distribution, stresses, wire tension for different wire speeds and a desired coating thickness. A full parametric study is undertaken for the dimensionless power-law index (in the case of pseudoplasticity) and the dimensionless yield stress (in the case of viscoplasticity). Pseudoplasticity leads to reduced pressure drop while viscoplasticity leads to increased pressure drop as a dimensionless Bingham number increases. All engineering quantities, given in a dimensionless form, increase substantially with the departure from the Newtonian values for viscoplastic materials. Although LAT gives good predictions for the engineering quantities, it produces misleading results for the yielded/unyielded regions, as evidenced by the full 2-D FEM analysis of the process.
(11-8-P) Poster Session

The Effect of Fiber-Glass Content on the Rheology of Semiconcentrated Polymer Composites

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Fiber-glass is used within a polymeric matrix to produce composites of enhanced properties and strength. The rheology of the composite depends on the fraction of fibers and their characteristics. Experimental data for fiber-reinforced composites are collected with the purpose of measuring and predicting their viscoelastic behaviour. Dynamic data ($G'$ and $G''$) and steady-state data (shear viscosity and normal stresses) are collected for fiber-filled viscoelastic media, namely polypropylene, polycarbonate, and nylon-66, for different sized fibers and different volume fractions. A constitutive equation is also used to describe the steady-state and dynamic rheological properties of fiber-filled viscoelastic media. This is an integral equation of the K-BKZ type, suitable for polymer solutions and melts. The contribution to the total stress of the composite due to the presence of fibers is accounted for by the extra stress tensor proposed by Dinh and Armstrong for fiber-filled Newtonian liquids. Steady-state predictions of the constitutive equation are compared with experimental measurements. The constitutive equation predicts qualitatively the rheological properties of fiber-filled viscoelastic media.

Notes:
The objective of this paper is to perform numerical calculations for non-uniform steady flows of polymer solutions and melts. The numerical simulation based on constitutive equation has the form of the Vinogradov phenomenological equation, which was modified by Pokrovskii and Pyshnograi. For the numerical computations used the SMAC method. The numerical study fluid flows through contractions, flows with free surface and extrudate swell. The results numerical simulations, experimental data and numerical results with other constitutive equation models are compared.

Notes:
A Structural Model for Thixotropic Fluids Exhibiting Yield Stress

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The elasto-thixotropic structural model of Fang and Tiu [Int. J. Eng. Fluid Mech. 3, 148-174 (1990)] is modified to describe the behavior of thixotropic fluids exhibiting yield stress. The effects of the yield stress and the kinetic orders of structural aggregation and segregation on the stress response in both formation and relaxation experiments is examined. Preliminary comparisons with experimental data on a model soft colloidal suspension are also presented.

Notes:
Long Chain Branches and the Rheology of Polypropylene

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Polypropylene (PP) is the most important commodity plastic. Most commercial PP grades, however, are not used directly in thermoforming or foaming processes because they have very low melt strength. In order to improve this property, long chain branches (LCB) are added on the (initially linear) chains. This can be simply done in extrusion by using reactive modification with specific peroxydicarbonates (PODIC).¹ LCB is controlled by the type and the amount of the PODIC, the molecular weight and the molecular weight distribution of the precursor.² The branched PP shows distinct strain hardening, something which is absent from the linear melt; the strain hardening is responsible for the considerable improvement of the melt strength. The zero shear viscosity and the elasticity, as indicated by the relaxation spectrum, also increase with the number of branches per molecule, \( B_n \). Two models are applied to describe the strain hardening of viscosity in the course of elongation: an extension of the Lodge Rubber-like liquid ³ that includes a damping function, and a modification of Wagner’s Molecular Stress Function theory.⁴ The model parameters are found to vary systematically with \( B_n \) and their values can be used as a measure of the degree of long chain branching. Consequently, fluidity, elasticity, strain hardening and melt strength are all related to \( B_n \). The processability in foaming and thermoforming processes improves with branching and shows an optimum, beyond which higher degrees of long chain branching appear not to help any further.


Notes:
Slip is exhibited by polymer melts in flows through processing equipment. Our previous work has documented the onset of slip for both linear (HDPE) and branched polymers (LDPE), and the alleviation of this phenomenon by using a judicious amount of processing aids. Recent experiments on a branched PP melt in a 10:1 axisymmetric contraction have demonstrated the onset of slip even for relatively low flow rates. The experimental device allows for the determination of slip velocity, while video processing of the flow shows the formation of vortices and their diminution with increasing flow rate due to slip. Numerical simulations using a multimode K-BKZ viscoelastic and a purely viscous (Cross) model – both of them incorporating a nonlinear slip law – were used to predict the flow kinematics and dynamics. It was found that the numerical predictions do agree well with the experimental results for the velocity profiles, vortex formation, and excess pressure losses through the contraction. The effect of adding processing aids does not affect the rheology but enhances slip at the polymer-wall interface, thus reducing the pressure drop and producing smooth extrudates. It is suggested that such experiments (visualisation of entrance flow) can be useful in evaluating the validity of constitutive equations and slip laws in the flow of polymer melts through processing equipment.
Human blood exhibits pronounced viscoelastic behaviour primarily caused by the structural element, the red blood cell (RBC) through its aggregation and deformability. It has been widely recognised that systemic arterial blood pressure depends on both cardiac output and on total peripheral resistance. The peripheral resistance is affected by haemorheology as well as the geometry of the arteries and capillaries. The aim of this project was to carry out quantitative rheometric characterisation of blood samples (including two ovo-vegetarian samples) in smooth and roughened rheometric tool surfaces in order to identify possible differences. Rheometric characterisations were performed using an ARES rheometer with a single and double wall Couette geometry. The aggregation state and deformability of RBCs under flow conditions were determined using Linkam optical counter rotational shearing cell with OLYMPUS microscope (50X lens) and PULNiX fast-capture digital camera for various shear rates. By closely examining the correlation between haemorheological properties and the structural evolution of the RBCs including deformation, aggregation and possible formation of shear bands in strong shear flow, we found that red blood cell aggregations and deformability are the main causes of the viscoelastic behaviour of blood and that factors like diet may significantly influence its flow behaviour. Testing blood in roughened rheometric tools revealed significant differences in low shear rates.

Notes:
An amphiphilic triblock copolymer, with hydrophilic mid-block and hydrophobic end-blocks, was synthesized by using group transfer polymerization GTP. The mid-block consists of 2-(dimethylamino)ethyl methacrylate DMAEMA, while the end blocks consist of methyl methacrylate MMA. This triblock copolymer tends to self-associate when it is dissolved in water, which is a good solvent for the PDMAEMA block and poor for the PMMA blocks. Thus the PMMA segments form compact cores, which are interconnected with PDMAEMA chains. These PMMA aggregates play the role of reversible cross-links between different copolymer chains.

In low concentration aqueous solutions, many different copolymer chains associate and form clusters of finite size, while in higher concentrations an infinite physical network is formed. This gel is soft (very viscous liquid) when the concentration is within the range 0.1 till 0.6 %w/v. When the copolymer concentration becomes higher than 1 %w/v a hard (solid like) gel is formed, which exhibit elastic behavior. Viscosity of such solutions presents four different regions when it is studied as a function of the applied stress. First there is a Newtonian plateau and then a dramatic decrease of the viscosity about four orders of magnitude. The high zero viscosity speaks for the formation of an infinite reversible network, which rapidly breaks down upon shearing. The infinite network brake into finite size clusters (microgels) which are able to flow giving much lower viscosity. The physical microgels are stable in a certain stress region and a second Newtonian plateau (third region) was observed. Finally, in the forth region a shear thinning effect can be seen which could be attributed to a further disruption of the microgels.

The critical gelation concentration was observed at 0.1 wt% above of which viscosity raises dramatically about nine orders of magnitude revealing that this polymer exhibits extremely high thickening efficiency.

The relaxation times seems to be very high showing that the life time of the physical bonds of the network is higher than the experimental time classifying these gels as strong physical gels.

Notes:
The Change of Rheological Properties of Waste Polymers in Thermal Degradation

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The reduction of the large amount of waste polymers (e.g. plastics) is more and more important from environmental aspects and furthermore their sufficient utilization is still an unsolved problem. Chemical recycling of waste polymers is one of the possible routes of their utilization. In this study the rheological properties of waste polymers (polyethylene, polypropylene) in thermal degradation process were investigated. The degradation of waste polyolefins was carried out in a horizontal tube reactor at 500-550 °C. Residues of distillation of wax-like products obtained by thermal cracking were studied. Some rheological properties e.g. melt-flow index (MFI), dynamic viscosity, shear rate, density, etc. were measured. It was found that the rheological properties of products obtained in thermal degradation of various plastics significantly changed during degradation. Considerable decrease of the average molecular weight and diminish of plastic range was noticed in thermal cracking. Relationships between the process parameters and the rheological properties of cracking residue were determined.

Notes:
Polyamide 6: pressure drop estimation through wire mesh filters and spinnerets

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Polyamide 6 is a polymer whose properties strongly depend on polycondensation reactions and its thermal history. Despite its wide use and a quite long presence on the market, still very few data may be found in literature for describing polyamides rheological behaviour. The main reason is that polymer properties change in time (in particular the molecular weight) when moisture content is not equal to the reaction's equilibrium value.

Rheological properties of an industrial Polyamide6, used for textile applications, have been studied using a rotational and a capillary rheometer. The polymer melt has been investigated at different temperatures and different moisture contents. For the examination with capillary rheometer different capillary dimensions were used. Measurements under oscillatory shear conditions were performed by rotational rheometer. The material functions were analyzed by an integral constitutive equation of the K-BKZ type with a PSM damping function, which describes the rheological behaviour of the polymer in many types of deformation. Polycondensation kinetics has been analyzed in terms of changes of complex viscosity and dynamic moduli with time.

The aim of the experiments and simulations were to gather all necessary material properties for evaluation of pressure drop values for polymer flow through two particular geometries: spinnerets having low L/D values (2-5) and wire mesh filters having openings in the range 10-100 µm. These two elements are commonly found in all spinning productions just before the extrusion. When they are properly combined, problems in production like frequent filtering elements replacement and spun-yarn breaks could be prevented.

Notes:
(7-O) Monday 12:20-12:40 Armonia 1

Observation of shear banding in ultrasoft colloidal gels

William M. Holmes,1 Paul T. Callaghan,1 Dimitris Vlassopoulos,2,3 Jacques Roovers4

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We explore the nonlinear rheological response of a soft gel formed by a crowded colloidal star polymer and focus on the occurrence of a stress plateau in its flow curve. With the aid of NMR velocimetry we attribute this plateau to shear banding and in particular to layer fluctuations across the gap of the measurement Couette device. Our velocity profile analysis supports a proposed three-states model of such fluctuations, according to which near the inner moving wall fluctuations are enhanced and relate to low viscosity liquid state, near the outer immobile wall an homogeneous high viscosity state of overlapping stars (forming some clusters) exists and the intermittent region is characterized by a jammed state (analogous to cages in colloidal glasses). These findings bring analogies to other classes of soft matter (such as surfactant micelles) exhibiting shear banding.

Notes:
Rearrangements and Yielding in Glasses of Hard and Soft Colloids

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The rheology and microscopic particle rearrangements during yielding of hard and soft sphere colloidal glasses was studied by a combination of conventional rheology and Light Scattering Echo (LS Echo). Creep and recovery measurements and dynamic strain sweeps showed that glasses of hard PMMA particles can tolerate surprisingly large strains, up to at least 15%, before yielding irreversibly. LS Echo verified that within their cage particles move reversibly at least up to such a strain. Such a behaviour was attributed to ‘cage elasticity’, the ability of a particle and its neighbours to retain their relative positions within the cage under quite large distortion.

To access the effect of interparticle interactions we also studied soft thermoreversible microgel particles where the interactions and the effective volume fraction may be tuned by temperature. In contrast to hard spheres the yielding does not depend upon the frequency, but only on the strain amplitude. We demonstrate that the drop of the echoes versus strain is related with the slow relaxation mode at rest. We then propose a physical picture to explain the shear thinning mechanism: the free diffusion at rest of ‘holes’ or ‘dynamical heterogeneities’ is replaced by forced motion under strain.

Notes:
Rheology of model branched polymers

M. Kapnistos$^{1,2}$, D. Vlassopoulos$^{1,3}$, L. G. Leal$^2$, J. Roovers$^4$

$^1$FORTH, Institute of Electronic Structure & Laser, Heraklion, Crete, Greece.
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Rheological and processing properties of entangled polymers have always been of great scientific and industrial interest. Different architectures and degree of branching substantially affect their mechanical properties.

We have measured the complete frequency spectrum of model linear and star combs. The Milner – Mcleish model with the tube dilution and arm relaxation, assumes a hierarchy of motions in branched systems. Using this model with appropriate modifications we can capture the relaxation of polymers of different chemistry and characteristics.

We have found that the idea works very well for different architectures and a frequency range of many decades. This gives confidence for the elucidation of the physics governing the motional mechanics of branched polymers.

Notes:
Rheological Properties of Controlled-Rheology Metallocene Polypropylenes

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Controlled-rheology polypropylenes (CR-PP) were produced in a Haake Rheocord 90 torque rheometer by using two metallocene grade polypropylenes (mPP) from Targor. Experiments were performed at 180 °C using various concentrations of Lupersol-101 peroxide. The molecular characteristics of the products were measured by high temperature size exclusion chromatography (SEC) and crystallization analysis fractionation (CRYSTAF). The rheological properties of these materials were measured in a Kayness Galaxy V capillary rheometer and in an AR2000 TAI parallel plate rheometer. The results showed that the polydispersity index (PDI) values of the molecular weight distributions (MWD) do not vary significantly in contrast with results for CR-PP obtained from conventional Ziegler-Natta (Z-N) resins. The linear viscoelastic properties were analyzed by using various rheological polydispersity measures and showed the potential formation of branching due to existing unsaturation in the virgin resins. An attempt was made to correlate these rheological polydispersity measures to SEC data and to highlight the differences between Z-N and mPP based CR-PPs.

Notes:
STRUCTURE AND DYNAMICS
OF POLYMERS
The Rheology of a Physical Gel Formed by Double Hydrophilic Triblock Copolymers

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Double-hydrophilic block copolymers are a new class of functional polymers of rapidly increasing importance with unique and fascinating properties. The combination of ionogenic monomers leads to block copolymers bearing oppositely charged blocks namely block polyampholytes. In this communication we present the various possibilities of an asymmetric ABA polyampholyte to form a variety of transient macromolecular assemblies by switching the solution pH, temperature, ionic strength and dielectric constant. More precisely, PAA$_{134}$ -P2VP$_{628}$ -PAA$_{134}$ exhibits different nano-structures in water such as spherical particles surrounding by negatively charged moieties, a stimuli responsive physical network and head to tail associates. All these assemblies are reversible adapting their structure by switching the environment conditions. Emphasis will be given to the rheological properties of a transient gel, which is formed by intermolecular electrostatic interactions, an unusual type of physical cross linking. Similarities and differences with a gel formed by hydrophobic interactions (PtBA$_{134}$ -P2VP$_{628}$ -PtBA$_{134}$/water) will be presented and discussed.

Notes:
Rheology and Morphology of Associative Telechelic Polyelectrolytes

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A poly(methyl methacrylate)-b-poly(2vinyl pyridine)-b-poly(methyl methacrylate) (PMMA-P2VP-PMMA), ABA triblock copolymer was synthesized by anionic polymerization with sequential addition of the monomers and investigated in aqueous media.

At low pH, P2VP is protonated and therefore is transformed to a cationic polyelectrolyte exhibiting hydrophilic behavior. Provided that P2VP is end-capped by very short hydrophobic PMMA blocks, the system is characterized as “telechelic” associative polyelectrolyte. This polymer self assembles in dilute aqueous solutions, forming flower-like micelles, as revealed by Static and Dynamic Light Scattering measurements and confirmed by Atomic Force Microscopy.

At higher concentrations (still in the dilute unentangled regime) a transient network is formed through transformation of the micellar structure as revealed by AFM. Steady state, oscillatory shear and creep experiments were performed to study the viscoelastic properties of the system. This kind of Physical gels exhibits unique properties due to the simple architecture of the macromolecule, and mainly to the stretched conformation of the protonated P2VP bridging chains.

Notes:
(17-O) Monday 15:40-16:00 Armonia 1

**Analysis of Shear-Induced Nematic-Isotropic Transition in Side-Chain Liquid Crystal Polymer**

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Despite the numerous information collected on viscoelastic properties of bulk polymers [1], very few studies reports on flow induced phase transitions. Liquid-crystal polymer (LCP) are composed of a polymer chain onto the sides of which mesogenic molecules have been grafted using a flexible chain called spacer.

![Schematic representation of side-chain liquid crystal polymer](image)

Recent studies have revealed that LCP melts exhibit strong non-linear deformations by shearing the material above a critical velocity gradient in the non-mesomorphic state (isotropic) [2]. This transition is the thermotropic equivalent of the well-known shear-induced nematic phase of wormlike micellar solution, extensively studied since 1994 [3].

For the first time, a comparative analysis of oscillatory birefringence flow and viscoelastic oscillatory shear experiments is carried out.

We have demonstrated that shear induced phase transitions can be inferred from a coupling neither with pretransitional fluctuations nor viscoelastic terminal time leading to consider longer time scales.

Recently, rheological measurements in the isotropic state revealed a low frequency elastic plateau. We tried to correlate the existence of this low-frequency plateau with the caracteristic time scale of the shear induced nematic phase [4].

R.H. Colby, J.R.Gillmor, Liquid Crystals, **13** (1993) 233;

Notes:
The Structure-Property-Relationship that is used the most is the Mark-Houwink-Sakurada relationship. This relationship that correlates the molar mass of a polymer with its intrinsic viscosity is well tabulated and easy accessible. (Brandrup and Immergut 1999).

More practical applications of Structure-Property-Relationships is the \( \eta_0 - M \) relationship that allows for a direct determination and calculation of the thickening properties of a polymer melt or highly concentrated solution (Bird, Armstrong et al. 1977). However, most technical applications of polymer solutions are in the semidilute concentration range where this simple \( \eta_0 - M \) relationship does not hold anymore (Grigorescu and Kulicke 2000). For a determination of the viscosity in this concentration range, the semi-empirical \( \eta_0 - [\eta] - c \) relationship can be established that holds even for systems where an exact determination of the molar mass is not possible (Clasen and Kulicke 2001). We have established these relationships for several different polymer-solvent systems, from polystyrene standards and cellulose derivatives to complex fermentation polymers.

These relationships can be expanded to the non-Newtonian flow regime (\( \eta = f(\gamma) \)) by introducing a Structure-Property-Relationship for the longest relaxation time via the \( \lambda - [\eta] - c \) relationship or the \( N_1 - [\eta] - c \) relationship to capture the elastic response of a polymer solution. These relationships allow for a complete formulation of the flowcurve in dependence of the molecular parameters. The advantage of these Structure-Property-Relationships lies in their simplicity and easy applicability for relevant problems.


Crumb rubbers - made from used tyres - were used to prepare bituminous compositions for road construction. A major problem in application is too high (it have to be pumpable) or too low (too much bleeding) viscosity of the binder. It was found that the viscosity of the rubberbitumens (RB) were changing in storage, depending on the preparation parameters and base materials. Dynamic viscosities were studied by using a rotating type of viscosimeter. Compositions proved, that the rubber modified bitumens were applicable at 135 °C, and at 180 °C they were pumpable. The effect of the size of crumb rubbers was also determined.

Notes:
Performance of Asphalt Rubber Versus Shear Stress

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The available European bitumen standards did not contain any requirements of measuring shear stresses of bituminous binders. However the upper layer of the asphalt have to be as resistant as possible, because about 85% of effects are shear stresses.

Crumb rubber modified bitumen compositions were prepared with different methods, from various base materials and their flow properties were studied by using a Brookfield viscosimeter. A new and fast test method was developed to characterize elastic relaxation. Results obtained by this test carried out within an appropriate temperature range suitably characterized the efficiency of the modification with crumb rubbers. So it was used for studying the performance of asphaltrubber. Results were evaluated by using a rheological model.

Notes:
We present measurements of collective concentration fluctuations in chemically end grafted polystyrene brushes swollen in solvents of various quality as probed by evanescent wave dynamic light scattering. [1] Two PS-brushes with a dried thickness of 8 nm (short) and 95 nm (thick) and molecular weight $M=10^6$ were used. 

In the good solvent (dioxane) regime, $C(q,t)$ (Fig. 2a) can be represented by single exponential decay with a diffusive relaxation rate $\Gamma(q)=D_c q^2$ (inset of Fig. 2a; $D_c=3.8 \times 10^{-7} \text{ cm}^2/\text{s}$ for the thick and $(2.8\pm0.5) \times 10^{-7} \text{ cm}^2/\text{s}$ for the short brush). Theoretically [3], $D_c=kT/6\pi\eta\xi$ describes the longitudinal mode of a monodisperse step-like brush with correlation length $\xi=d$ where $\eta$ is the solvent viscosity. The relation $D_s \propto 1/\eta$ was confirmed for the same brush in two different good solvents whereas the low value of $\xi$ in these polydisperse brushes is of the order of grafting distance d. 

When the solvent environment changes from marginal to poor, $C(q,t)$ (Fig. 2b) reveals strong effects as compared to the smooth variation of the density profile. In cyclohexane ($T_\Theta=35^\circ C$) only above $50^\circ C$ approaches the situation in dioxane. At lower temperatures $C(q,t)$ is clearly a two step decay where the fast decay is still assigned to the cooperative diffusion (albeit slower than in the good solvent regime) and the slow non-exponential process. The slowing down of the characteristic time together with the increase of the associated intensity with decreasing temperature are the characteristics of the slow mode that probably relates to the self-diffusion of clusters in analogy to gels. Based on a current documentation of the structural behavior of brushes, this rich and complex dynamic structure is rather unexpected.


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Notes:
We report on a telechelic associative polyelectrolyte (polystyrene - b – poly(sodium acrylate) - b – poly(-n-butylnmethacrylate)) in aqueous solutions that self-assembles into starlike flowers in the dilute regime and develops a fully connected physical network above some threshold concentration (0.1 wt %). The specificity of this system is that the hydrophobic domains are connected by stretched polyelectrolyte chains (due to electrostatic repulsions) that form a strong physical network. The aim of this work is to determine the influence of various external factors (salt, degree of neutralization and methanol addition) on the rheological properties, with the aim of achieving molecular control of the flow behavior in such amphiphilic systems. Due to the polyelectrolyte character of the bridging chains, the physical gel is sensitive to external stimuli, i.e. small changes in the external conditions such as pH, ionic strength and solvent quality can induce significant changes on the rheological profile of the solutions. Another important factor that influence remarkable the rheological properties of the solution is the copolymer architecture. To this respect a series of ABC copolymers, differing in the length of the C block was examined. An unusual behavior was observed, showing that an increase of the C block length, results to a decrease of the relaxation time of the network.

Finally with the aid of dynamic light scattering and scanning electron microscopy the association mechanism was studied in order to better understand the rheological behavior of the system.

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