CONEX Mid-term report
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1. Conditions for creating thin liquid layers a t the contact surface of two other liquids

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The design of new technologies, making it possible to manufacture the nano - structured materials is one of the most important tasks of the contemporary materials science. The technologies, utilizing the emulsion droplets as templates for producing nano - structures out of solid particles, suspended in the liquid phase, seem very promising; work on developing such technologies is progressing fast [1]. It seems however conceivable, that even smaller structures could be obtained, if instead of solid particles a liquid film was utilized. In the present research we investigate the conditions necessary for producing a liquid film at the interface of two non-mixing liquids.

To simulate the behaviour of the three liquids the Molecular Dynamics simulation technique is applied using the program MOLDY [2]. Since, at the molecular level, the behaviour of the liquids depends, in the first place, on the interaction potentials between the molecules [3], we look for the combination of the interaction potentials, which might produce the required liquid film at the surfaces of the emulsion droplets. We perform a number of simulation runs, assuming different interaction potentials. The liquids used for simulations are actually model liquids with desired properties, not the real ones. Still, the information, relevant for the technology, is obtained in this way. Having the information on the required interaction potentials one can look for the real liquids, fulfilling these requirements.

The aim of the preliminary calculations was to simulate formation of the liquid droplet in a vacuum and in another, non-mixing liquid. We placed a number of liquid molecules in a rectangular lattice and allowed them to move freely with velocities, corresponding to a prescribed temperature.

At the beginning we tried, for simplicity, to use the Simplified Water Model (SWM) . A liquid of spherically symmetric molecules, interacting with each other only through the Lennard . Jones potential, the same as that for water. Such molecules, placed in a vacuum, spread uniformly in the whole simulation domain. To form a well . defined droplet it was necessary to use more realistic model of the water molecule (TIPS2, [4]), taking into account the electrostatic dipole interactions. Such interactions, on top of the Lennard Jones potential, are the most important factor influencing the behaviour of water.

The initial configuration of the water molecules, taken for simulation, was actually very far from spherical. This produced oscillations of the shape of the droplet, visible for certain amount of time. The frequency of these oscillations was comparable to the mean molecular speed divided by the diameter of the droplet.

To simulate the formation of a liquid droplet, immersed in another, non-mixing liquid, we first tried the SWM molecules, however without electrostatic dipole interactions certain amount of mixing was always present. To produce a liquid droplet successfully we had to immerse the SWM liquid, (which now could represent oil) in water described by the TIPS2 model.

Results of the simulation are quite reasonable allowing to analyse physics of the two or three phase interactions of emulsions. Our present task is to perform simulations using larger

number of molecules and to introduce interactions potential for the mixtures used in the CONEX experiments.

References

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2. High-frequency linear viscosity of emulsions composed of two viscoelastic fluids

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We consider high-frequency linear viscoelastic response of an emulsion composed of viscoelastic drops suspended in another viscoelastic fluid. Typical examples of such systems include polymer blends and emulsions of polymeric solutions. Due to importance of viscoelastic fluids in chemical, pharmacological, and food industries, viscoelastic emulsion properties have been intensively studied theoretically and experimentally. For moderate drop volume fractions and viscosity ratios between the drop and continuous fluids simple analytical expressions for the effective viscosity of an emulsion have been developed. However, such approximations are inaccurate at high volume fractions of highly viscous drops.

In this work we focus on the frequency regime where the timescale associated with the frequency of the imposed flow is much smaller than the drop capillary relaxation time.
In this frequency regime the effect of the interfacial tension on drop dynamics can be neglected; yet, the system has nontrivial viscoelastic response due to the viscoelasticity of the component fluids. The drop deformation is assumed to be small at all times, and the response of the component fluids is linear.

Under the high-frequency assumption and small-deformation conditions the drops behave as nearly-spherical viscoelastic fluid blobs with complex viscosity ratio. Due to the long capillary relaxation time the drops passively react to the imposed flow. We have derived explicit expressions for the function in terms of the spectrum of the operators characterizing hydrodynamic interactions of spherical drops. The expressions have been evaluated numerically for different volume fractions of randomly distributed drops. Our results allow to calculate the effective viscosity of an emulsion with real values of the viscosity ratio.

Using Bergman spectral representation, we have developed a complete description of high-frequency effective viscosity for an emulsion of viscoelastic fluids. We have evaluated numerically the spectral density and the coefficients of the continued-fraction expansion. With only several expansion levels, the continued fraction provides a very accurate analytical

representation of the effective viscosity. The work is underway to extend this approach to the frequency regime, where the interfacial-tension effects are important.

Performed analysis can be used to predict behaviour of emulsions applied in the CONEX experiments.

References

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