

# Plenary Lectures

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## Mastering colloids and interfaces: key to innovation in a convergent research model

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Research in the fields of energy, environment, water and human health currently is too much disconnected, which is suboptimal since all rely on very similar physicochemical concepts and research approaches at the molecular scale and they could benefit from shared scientific knowledge. Current inter- and transdisciplinary collaboration of mono-disciplinary experts meets with some success, but such collaboration is slow and may be insufficient to solve the global problems humanity is faced with.

Convergence is a new paradigm proposed in 2011 by MIT [1] and later adopted by the NSF USA, moving beyond the current inter- and transdisciplinary collaboration models, as it proposes truly transdisciplinary integration of Life & Health, Physics & Chemistry, Engineering Sciences to accelerate technological progress in challenging areas of concern to mankind, like sustainable energy, food, climate, health and water.”

In the talk a new initiative at KU Leuven, the “Convergent Research Fund’ fostering the convergent research idea is presented. Knowledge on how molecules organize themselves at interfaces, how they are transported through membranes and pores, and adsorbed and converted on catalysts is gained from advanced Nuclear Magnetic Resonance spectroscopy. The first successes of convergent research at KU Leuven are presented: in sustainable energy, the development of hydrogen panels, trapping water from atmospheric air and converting it to hydrogen and oxygen gas using solar energy, in chemical manufacturing, altering water properties by hydrophobic interaction to make it less polar and fit for dissolving organic molecules to replace organic solvents; in the field of food production, original concepts for off flavor abatement in aquaculture and fish farming are presented, and in the pharmaceutical area, enhanced bioavailability of poorly soluble drugs by using silica-based drug delivery. It will be explained how the activities are being expanded to the biomedical area.

### References

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## Flexible colloidal structures

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Many functional (bio-)molecules possess structural flexibility which provides functional properties and a more complex phase behavior. For example, different conformations may enhance or inhibit a protein's activity in allosteric regulation or enable the catalytic activity of enzymes. Colloidal structures currently lack this property, restricting their use as building blocks in reconfigurable materials and model systems. In this talk, I will introduce colloidal particles with surface-mobile DNA linkers [1,2] – so called colloidal joints [3]- that enable the formation of strong and specific hinging bonds. I will describe how this bond mobility affects their diffusive and self-assembly behavior and demonstrate the assembly of a variety of flexible structures. In particular, I will discuss the formation of colloidal molecules with tunable flexibility and shape in high-yields. These novel colloidal building blocks give access to a new class of materials with great potential in shape-shifting systems, actuators and colloidal robots.

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## Mesoporous silica designs tailored for nanomedicine

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One of the main objectives in the development of nanomedicines is to obtain delivery platforms for efficient and targeted delivery of drugs and/or imaging agents to improve therapeutic efficacy, reduce side effects and increase diagnostic sensitivity. A material class that has been recognized for its controllable properties on many levels, which could facilitate the development of such systems, is mesoporous silica nanoparticles (MSNs). Associated traits behind this success are the modularity combined with the vast surface functionalization approaches adoptable for MSNs. Here, the chemical, structural, and textural robustness of the architecture of this inorganic material platform can be readily fine-tuned with the aid of responsive, biocompatible and flexible organic modifications to form multifunctional hybrid materials (Figure 1). Moreover, the four distinct domains that can be independently functionalized,<sup>[1]</sup> i.e. the silica framework, the particle surface, the pore walls, and the inner space in the case of hollow MSNs; endows this platform with especially flexible design possibilities.

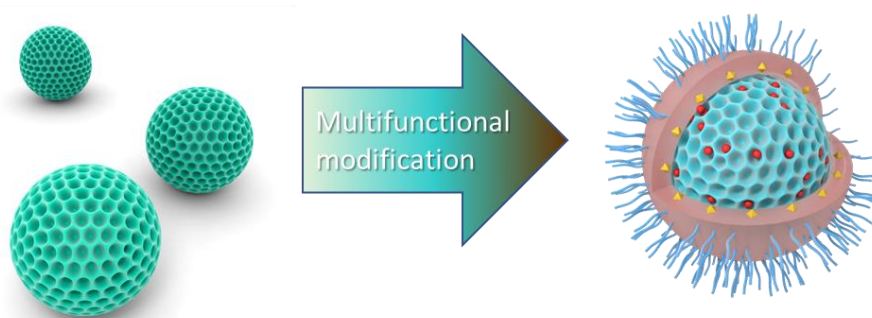


Figure 1: Using inorganic porous nanoparticles as a platform for further modification allows for a multitude of functions to be incorporated for a vast array of applications within nanomedicine.

Silica is also the most widely used coating for other inorganic nanoparticles, since it is biocompatible, allows for easy further functionalization, and efficiently protects the core material. If the cores are coated with mesoporous silica, the porous shell can also be utilized for incorporation of active molecules such as drugs or molecular imaging agents, creating e.g. multimodal fluorescent-magnetic nanocomposites.<sup>[2]</sup> Inorganic nanostructures are readily utilized as biomedical imaging probes due to their inherent detectability by a variety of different imaging modalities, e.g. nanodiamonds for optical imaging<sup>[3]</sup> and superparamagnetic iron oxides, SPIONs, for magnetic resonance imaging (MRI). These materials are thus frequently coated with porous or non-porous silica layers in the design of multifunctional core-shell materials.

This Plenary Lecture will outline design aspects that emphasize the utilization of MSNs as a versatile platform for nanomedicine development, covering the whole range from materials design and formulation with different active agents, to *in vitro* cellular evaluations and *in vivo* therapeutic delivery and detection.

### References

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## Colloidal Forces

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Numerous phenomena are governed by forces between colloidal particles. Examples include stability of colloidal suspensions, their rheology, or formation of colloidal crystals. The importance of such interaction forces has been appreciated for a long time, but accurate measurements of such forces remained a considerable challenge until quite recently. Presently, however, various techniques are becoming available for direct force measurements. In my view, the colloidal probe technique based on the atomic force microscope (AFM) is emerging as the most promising and versatile one.

The present talk will discuss the use of this technique to measure forces between colloidal particles in aqueous systems. In particular, I will demonstrate that interactions between similar as well as dissimilar particles are principally controlled by electrostatic double-layer and van der Waals forces as already suggested by the classical theory of Derjaguin, Landau, Verwey and Overbeek (DLVO). The talk will further highlight the nature of forces acting between charged colloidal particles in the presence of multivalent ions and polyelectrolytes. While forces in these systems remain DLVO-like and are mainly controlled by charge reversal, additional attractive non-DLVO forces come into play. Additionally, two novel phenomena will be highlighted as well. First, charge regulation effects can become very important for dissimilar particles. Second, double layer forces can become strongly non-exponential in the presence of multivalent coions or like-charged polyelectrolytes.

## **Cryo-EM of Complex Liquids: A Brief History and the State-of-the-Technology**

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It is well accepted in modern science and technology that direct-imaging on the supra-molecular scale is an essential part of full nanostructural characterization of any material system, including complex (nanostructured) liquids. For the latter, the main established tools are cryogenic-temperature transmission electron microscopy (cryo-TEM) and cryogenic-temperature scanning electron microscopy (cryo-SEM). Both techniques, referred to jointly as cryo-EM, involve ultra-fast cooling of the specimen, and its examination at cryogenic temperatures. That makes the specimens compatible with the microscopes, and arrests all perceptible motion during imaging. Another challenge is keeping the temperature and composition of the studied system unaltered during specimen preparation, until it is thermally fixed.

Cryo-EM has its roots in the late 1970s. Over the years many scientists and technologists contributed to the development of specimen preparation, improving the microscopes, improving imaging, and understanding the physics and chemistry of electron beam-specimen interaction. All that has led to better preservation of the specimens during preparation, transfer into the EM, and imaging, improved image resolution, and extension of the application of the methodology to a very wide range of the studied systems, from synthetic to biological, from the aqueous to the nonaqueous, even to systems based on strong acids.

In will start my presentation with a brief history of cryo-EM, especially as applied in the study of complex liquids. Then I will describe and demonstrate some the state-of-the-technology features of cryo-TEM and cryo-SEM that have made very strong impact on the field in recent years. In the last part of the talk I will present recent applications of modern cryo-EM in nano- and biotechnology.

## New approaches for surfactant control of foam and emulsion properties

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We have revealed that individual surfactants and their mixtures with other surfactants, polymers and/or particles can be used as powerful active agents for intelligent control of foam and emulsion properties. Four new approaches will be presented as case studies: (1) We showed that two different mechanisms, bulk gelation and Pickering stabilization, can be used for production of ceramic porous materials, with excellent thermal and mechanical properties, from foamed suspensions. These mechanisms lead to materials with different structure and properties, e.g. with open or closed pores. This approach can be applied to different types of solid particles by selecting appropriate surfactants. (2) We showed that one could use cosurfactants of different chain-lengths, various cationic polymers and counterions to modify the kinetics of surfactant adsorption, surface viscoelasticity, bubble-bubble attraction and the related foamability and foam rheological properties of the surfactant solutions. These studies have important implications for the home and personal care formulations. (3) We studied systematically a large series of natural surfactants, called “saponins”, which have high surface activity and are used industrially as foamers and emulsifiers. Various plant extracts were compared and wide diversity of surface properties was observed – some saponins exhibit extremely high surface viscoelasticity, while others have other beneficial properties. We showed that the high surface viscoelasticity of the triterpenoid saponins (which differ from the steroid ones) is due to strong hydrogen bonding between the sugar residues of the adsorbed saponin molecules and to tight packing of the rigid hydrophobic scaffolds of these molecules in their adsorption layers. These properties are very different from those of the conventional surfactants. (4) In collaboration with colleagues from the University of Cambridge, UK, we have observed and explained a range of unexpected and non-trivial phenomena in cooled alkane-in-water emulsions, including spontaneous drop self-shaping and drop self-bursting when the alkane drops undergo liquid-to-solid phase transition. Although the mechanisms of these fascinating phenomena are still debated, we have demonstrated how one could use and/or combine different long-chain surfactants to push the system into different desired scenarios.

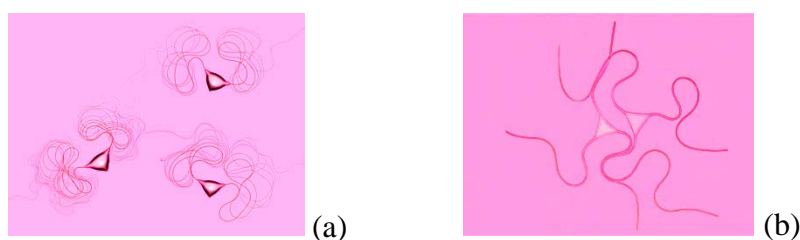


Figure 1: Examples of spontaneously formed oil drops with flagella obtained via cooling of (a)  $C_{15}$ -in-water and (b)  $C_{16}$ -in-water emulsions, stabilized by  $C_{16}EO_{20}$  nonionic surfactant (Brij 58).

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