Modelling hydrogels: building networks in the Mathematical Sciences

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Hydrogels have found an increasing number of uses in wide and varied fields since their first proposed application in contact lenses in the 1960s. Their large-swelling, shape-transforming, elastic and biocompatible properties have been exploited in medicine, soft robotics, agriculture, materials science and even heavy industry, bringing together researchers in the applied sciences and product development. Modelling these complicated behaviours requires insights at multiple scales, from the microscopic interactions between water and polymer molecules, thermodynamical considerations and also the bulk macroscopic poroelastic behaviour seen during swelling or deformation. Though there has been great progress on this front in recent years, many models are phenomenological and there are significant gaps in our knowledge. Furthermore, the most successful developments in recent years have combined insights from a number of different, sometimes disparate, fields, underlining the importance of collaboration and discussion in making progress. Below, three *grand challenges* with some key questions are summarised, highlighting some of today's key topics of discussion.

Living gels, living with gels: hydrogels in microbiology and medicine

Speaker: ***tba***

Gels grow upon immersion in water or other solvents, and can shrink as they lose water to their surroundings. This immediately invites comparison with a number of biological processes involving morphogenesis and development (embryo growth, formation of wrinkles on the brain, plant development…), as well as the dynamics of biofilms and tumour growth. Therefore, we can consider hydrogels as ideal proxy materials for cell colonies, biofilms or developing tissues, and insights from continuum-mechanical modelling can be applied to understanding systems of cells or microorganisms *ex vivo*. Furthermore, some of the most prevalent applications of hydrogels are found in biomedical contexts, leveraging the biocompatibility and softness of these materials in implants and tissue dressings, or in slow-release capsules for the targeted delivery of drugs in the digestive system or bloodstream. More recently, research has focused on the use of hydrogel scaffolds for the growth of synthetic organs and on crafting implantable devices that are soft and compatible with a variety of tissue types, both applications which require a detailed understanding of the physical properties of the gel and how it may evolve and perform over long timescales, and insights from biologically-driven models for simple multicellular life could equally inform our own approaches to modelling gels.

Zohuriaan-Mehr *et al.* (2010) *Advances in non-hygienic applications of superabsorbent hydrogel materials* J Mater. Sci. 45:5711-5735
Ido *et al.* (2020) *Bacillus subtilis biofilms characterized as hydrogels. Insights on water uptake and water binding in biofilms* Soft Matter 16:6180-6190
Prince *et al.* (2020) *Biomimetic hydrogel supports initiation and growth of patient-derived breast tumor organoids* Nat. Commun. 13:1466

At the surface and through the pores: transport and interfacial behaviour

Speaker: ***tba***

The vast majority of applications of hydrogels involve the transport of water and solvents throughout the pore network. Swelling and drying require the accumulation and expulsion of water, respectively, and deforming the gel structure can also drive flows as water is squeezed from stressed to unstressed regions. Modelling the flow of water and its effects on the structure of the polymer scaffold is therefore a key problem that has been overlooked in many existing approaches, which simply assume a Darcy flow model with a single-phase fluid. In addition to seeking a comprehensive understanding of the behaviour of the interior of a gel, a complete model for hydrogel dynamics must also consider interfacial conditions. Generally, modelling the interfaces between gel and the surroundings poses more of a challenge than describing the interior. A sound understanding of the correct conditions to apply at gel-water, gel-solution or gel-air boundaries is vital to model swelling, drying and transport of solute in hydrogels, and is also of paramount importance when we consider gels that respond actively to their environment. Questions of the importance of elastocapillarity (surface tension in soft solids) and the nature of the surface boundary conditions or the microscopic-scale structure of the surface of a gel in air are of clear importance but have not yet been satisfactorily addressed by modellers.

Eddington & Beebe (2004) *Flow control with hydrogels* Adv. Drug Delivery Rev. 56:199-210
Wheeler & Stroock (2008) *The transpiration of water at negative pressures in a synthetic tree* Nature 455:208-212
Etzold *et al.* (2021) *Transpiration through hydrogels* J Fluid Mech. 925:A8
Style *et al.* (2017) *Elastocapillarity: Surface Tension and the Mechanics of Soft Solids* Annu. Rev. Cond. Matt. Phys. 8:99-118
Xu *et al.* (2024) *A theory of hydrogel mechanics that couples swelling and external flow* Soft Matter (accepted, 10.1039/d4sm00424h)

Feeling the surroundings: hydrogels as smart materials

Speaker: *tba*

In recent years, hydrogels that respond to external stimuli such as temperature, light, pH and the presence of various chemical species have found applications in soft robotics and microfluidics. Being able to tune the shape of a soft material as the surrounding environment changes allows for the creation of actuators, microfluidic valves and self-propulsive objects. With a sound and accurate description of the processes by which such materials do respond to stimuli, we can tune the specific timescales and extent to which the shape and composition will evolve, allowing us to more finely control the creation of smart devices at the desired scale. Modelling these gels requires us to couple established approaches for the behaviour of swelling soft matter with the physics governing the active properties, incorporating insights from continuum mechanics, thermodynamics and electrochemistry. We are currently at an exciting stage in the development of these materials as experimentalists begin to discover new applications for active superabsorbent gels, feeding in to a rapidly rising demand for modelling insights.

Chester & Anand (2011) *A thermo-mechanically coupled theory for fluid permeation in elastomeric materials: applications to thermally responsive gels* J Mech. Phys. Solids 59:1978-2006
Cai & Suo (2011) *Mechanics and chemical thermodynamics of phase transition in temperature-sensitive hydrogels* J Mech. Phys. Solids 59:2259-2278
Butler & Montenegro-Johnson (2022) *The swelling and shrinking of spherical thermos-responsive hydrogels* J Fluid Mech. 947:A11