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Nanotribology of charged polymer brushes[1]

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Polymers at surfaces, whose modern understanding may be traced back to early work by Sam Edwards[1], have become a paradigm for modification of surface properties, both as steric stabilizers and as remarkable boundary lubricants[2]. Charged polymer brushes are of particular interest, with both technological implications and especially biological relevance where most macromolecules are charged. In the context of biolubrication, relevant in areas from dry eye syndrome to osteoarthritis, charged polymer surface phases and their complexes with other macromolecules may play a central role. The hydration lubrication paradigm, where tenaciously-held yet fluid hydration shells surrounding ions or zwitterions serve as highly-efficient friction-reducing elements, has been invoked to understand the excellent lubrication provided both by ionized[3] and by zwitterionic[4] brushes. In this talk we describe recent advances in our understanding of the nanotribology of such charged brush systems. We consider interactions between charged end-grafted polymers, and how one may disentangle the steric from the electrostatic surface forces[5]. We examine the limits of lubrication by ionized brushes, both synthetic and of biological origins, and how highly-hydrated zwitterionic chains may provide extremely effective boundary lubrication[6]. Finally we describe how the lubrication of articular cartilage in the major joints, a tribosystem presenting some of the greatest challenges and opportunities, may be understood in terms of a supramolecular synergy between charged surface-attached polymers and zwitterionic groups[7]. 1. Dolan & Edwards, **Proc. Roy. Soc. A**, **337**, 509 (1974). 2. Klein et al. **Nature, 370**, 634 (1994). 3. Raviv et al., **Nature**, **425**, 163 (2003). 4. Chen et al., **Science**, **323** 1698 (2009). 5. Peretz et al., to be published. 6. Tairy et al., **Macromolecules**, **48**, 140 (2015). 7. Seror et al., **Nature Communications**, |6:6497 |(2015); Jahn et al., **Annual Reviews of Biomedical Engineering** (2016)

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