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Competitive specific ion effects: A neutron reflectometry study of thermoresponsive polymer brushes in mixed electrolytes

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Specific ion effects are phenomena that depend on the identity of ions present in a system, and not merely their valence or concentration. For example, the Hofmeister series orders ions on their ability to either stabilise (salting-in ions) or destabilise (salting-out ions) proteins and is vital in biochemistry.[1] In recent years, polymer brushes, which consist of end-tethered polymer chains to a substrate, have been used as exemplar systems to investigate specific ion effects.[2] The effective solvent quality influences the conformation of these brushes (collapsed or expanded), which is directly linked to application properties (e.g. switchable adhesion and self-cleaning). We have performed a significant body of work on brush conformation in single salt electrolytes.[2-4] However, our understanding of the influence that mixed electrolytes have on the behaviour of polymer brushes is currently limited, which is necessary for real-world applications. Here, we present the behaviour of poly(ethylene glycol) methyl ether methacrylate (POEGMA) brushes in a variety of both pure and mixed electrolytes, as studied with neutron reflectometry. Reflectometry allows for the extraction of volume fraction profiles, which can provide detailed information regarding the influence of ion specificity on polymer brush conformation. Consistent results were also obtained from other techniques, such as ellipsometry, which is used to track overall changes in brush thickness.

In the presence of electrolytes composed of ions from the same end of the Hofmeister series (salting-in and salting-in or salting-out and salting-out), a non-monotonic concentration-dependent influence of the two ions was observed. The specific ion effects imparted by two salting-in ions were dependent on the influence of the ions with the polymer chains. In contrast, the impact of two salting-out ions was dependent on the available solvent molecules. In the presence of electrolytes composed of ions from opposite ends of the Hofmeister series (salting-in and salting-out), ion behaviour was observed to be temperature-dependent.[3] Much can be gained by improving our knowledge of ion specificity and understanding the subtle structural changes of a brush are essential in order to unravel the dominant drivers behind specific ion effects.

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[2] T. J. Murdoch, B. A. Humphreys, J. D. Willott, S. W. Prescott, A. Nelson, G. B. Webber and E. J. Wanless, *J. Colloid Interface Sci.*, 2017, **490**, 869–878.

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