MAR12-2011-001155

Abstract for an Invited Paper
for the MAR12 Meeting of
the American Physical Society

Adhesion of hydrogels under water by hydrogen bonding: from molecular interactions to macroscopic adhesion[1]

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Hydrogels are an essential part of living organisms and are widely used in biotechnologies, health care and food science. Although swelling properties, cell adhesion on gel surfaces and gel elasticity have attracted much interest, macroscopic adhesion of hydrogels on solid surfaces in aqueous environment is much less well understood. We studied systematically and in aqueous environment, the reversible adhesion by hydrogen bonding of macroscopic model hydrogels of polydimethylacrylamide (PDMA) or of polyacrylamide (PAAm) on solid surfaces functionalized with polyacrylic acid (PAA) polymer brushes. The hydrogels were synthesized by free radical polymerization and the brushes were prepared by grafting polytertbutyl acrylate chains and converting them by pyrolisis into polyacrylic acid. A new adhesion tester based on the flat punch geometry was designed and used to control the contact area, contact time, contact pressure and debonding velocity of the gels from the surface while the samples were fully immersed in water. The adhesion tests were performed at different pH and temperatures and the modulus of the gel and grafting density and molecular weight of the brushes was varied. Macroscopic adhesion results were compared with phase diagrams in dilute solution to detect molecular interactions. While the PDMA/PAA pair behaved very similarly in solution and in macroscopic adhesion tests, the PAAm/PAA pair showed an unexpectedly high adhesion level relatively to its complexation ability in dilute solution. Surprisingly, time dependent experiments showed that the kinetics of H-bond formation and breakup at interfaces was very slow resulting in adhesion energies which were very dependent on contact time up to one hour of contact. At the molecular level, neutron reflectivity showed that the equilibrium brush conformation when in contact with the gels was more extended at pH2 (H-bonds activated) than at pH9 (H-bonds deactivated) and that a certain applied pressure was necessary to bring the brush and the gel in contact. These combined results suggest that the mechanisms of formation and breakup of interactions at surfaces are more complex and cannot be understood from bulk interactions in solution alone.

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[1] We gratefully acknowledge support from the French ANR: project AdhGel