

# SUMMARY

Standard molecular dynamics software uses sophisticated and realistic potential functions. However, the number of atoms in the simulated systems is seldom more than a few thousand. At the same time, specialised scientific codes have been developed, based on thermodynamic statistics that use more comparatively simple potentials. The original aspects of this PhD thesis lies in combining these advantages: big systems and realistic potentials where they are of crucial importance, close to interfaces.

The central thread of this work is the improvement and use of molecular dynamics techniques as applied to interfacial phenomena. This thesis reports a number of studies. Their main objectives are either to explain or validate experimental data or to improve our understanding of the observations. In this cycle lies the interest: experiments suggest simulations, simulations enhance experiments.

Molecular dynamics can also be of great help to theoreticians, the first study is a typical example. Very often, analytical theories are achieved only with help of approximations, whose validity is not always obvious. In our case, an analytical formulation for the spreading of polymers (the Growing Disk Model) is an approximation used to develop a complete theory for the kinetics of polymer adsorption. Numerical simulations justify the use of this approximation. In the same area, we study the spreading of polymer binary mixtures on top of a solid. Special attention is paid to the influence of preferential interaction between a given polymer type and the solid on the global kinetics of spreading.

Opposing monolayers of lipids which have nucleotides as the headgroups show unexpectedly long range attraction. The most likely explanation for those observations is the organisation of the monolayer molecules into oriented clusters. Using molecular dynamics simulations, we have been able not only to highlight the existence of domains but also to propose an experimental procedure to unambiguously identify the mechanism responsible for this specific interaction.

In electrokinetics the existence of the double layer is well established. Experience has shown that a part of the double layer remains stagnant in electrokinetic phenomena. The width of this "immobile layer" is of the order of molecular dimensions. Despite it's apparent immobility, there is ionic conduction in the stagnant layer which has to be accounted for. What are the origin and properties of this stagnant layer? How do ions move in it? These questions have now been answered. Viscosity computed as a tensor reveals a strong anisotropy between directions parallel and normal to the solid plane. The stagnant layer behaves as a two-dimensional gel, whose viscosity is higher than bulk phase, whilst the mobilities of ions in it are hardly impaired.

Finally we have studied, at the molecular scale, the structure of an interface between immiscible chain-like liquids under shear. We have been able to highlight the influence of both shear and molecular structure on dynamic properties such as interfacial tension. These results open new opportunities to improve our understanding of complex hydrodynamic problems such as those involved in industrial multi-layer coating.