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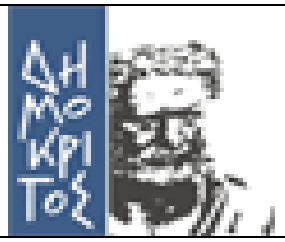

Research Directions

As of 2013, a major restructuring of the group took place in order to meet the recent needs of EU projects and the demands for a stronger collaboration with the industrial sector. Some old friends have gone to work in other projects and new ones appeared. We hope for the best in the forthcoming years with the new EU HORIZON framework. Most of the current research activities and directions are explained in the next paragraphs.

Continuous developments in computational infrastructure, on the one hand, and in the areas of nanomaterials, biomaterials, etc., on the other, bring the computational study of matter to the fore, with special emphasis on a multiscale approach that looks at all structural levels from the atomic and molecular through the mesoscopic to the macroscopic one. Our group contributes actively in this direction, especially in the fields of Molecular Modeling and thermodynamics of physico-chemical systems that interest practical applications.
The broad directions of our work comprise a) development of software applications with the ultimate aim of creating a unified multitask material modeling platform b) novel techniques and algorithms c) carrying out computations and specific case studies d) related scientific publications.
In terms of specific areas of research and development, the above can be categorized as follows:

1. Structure, dynamic and thermodynamic properties:
Molecular simulations are typically followed by a series of routine post-processing tasks to obtain the properties of interest. Depending on the case, these computations may concern the pair distribution functions of atoms, groups of atoms or molecules and their Fourier transformation that give valuable information about a system's structure; auto-correlation functions of velocities or vectorially expressed geometric quantities to describe its dynamics at the microscopic level; a range of thermodynamic properties, including density, compressibility, thermal expansion coefficient, elastic moduli, surface tension, or, in the case of coexisting phases (such as surfactant/water, oil/water systems etc), density profiles, interfacial tension etc. Currently, we are working on the development and constant update of an easy to use post-processing toolkit to cover a broad range of these tasks, including some advanced techniques that are not implemented in common molecular simulation packages.

2. Transport properties:
Diffusion can be a key process in industrial applications, materials engineering or natural and biological systems. Its knowledge and understanding is indispensable for theoretical studies as well as the design of practical applications. Our work in this area has to do with the prediction of *diffusion coefficients* in binary and multicomponent systems in the Fickian and Stefan-Maxwell framework; study of anisotropy of molecular diffusive motion by means of the diffusivity tensor; understanding of the microscopic mechanisms underlying mass transport processes (for instance, quantitative study of diffusive molecular jumps by means of a unique technique introduced by V. Raptis and T. Raptis and later, employed in modeling and experimental studies). In the same vein, other transport properties such as viscosity are or great interest when it comes to the study, from a rheological point of view, of processes involving fluids or polymer melts. In this area, we look at the use of both the Green-Kubo and Einstein formalisms to link dynamic behavior at the microscopic level with predictions of viscosity and other macroscopic properties.

3. Polymer Systems:
Polymers are a very important class of materials with numerous applications. Their study via molecular simulation can be challenging due to their notoriously long relaxation time scales. That is why the optimal choice of an initial structure is crucial to avoid the need for long equilibration computations. Building a realistic initial polymer configuration can be achieved through special "amorphous builders" which must guarantee the best entropic properties of such structures. We are working on a generic software product able to treat monodisperse and polydisperse structures for linear polymers and, prospectively, branched chains, dendritic molecules and other complex structures.
Using the same software libraries developed so far, we have been able to implement an efficient scheme to emulate an important class of polymerization reactions. The emulator, which is under continuous development, allows to study the factors that affect the degree of polymerization and the dispersity index of the resultant product by letting the user define certain input parameters that influence the final outcome.

4. Molecular Mechanics:
In combination with the amorphous builders, a generic optimization scheme may be required to assure that a configuration is energetically minimal. To this direction, we implement several schemes, which we interface with the "Merlin" package, a versatile minimization toolkit developed in the University of Ioannina, and its descendants "PANMIN" and "MEMPSODE" that implement global optimization algorithms. In cases of single molecule energy minimization, the method can effectively be used as a first stage for the design of optimal force-fields to be used later in Molecular Dynamics and Monte Carlo simulations.

5. Development of novel Force-Fields:
This is a very large topic where several methods are constantly under development worldwide. The development of so-called coarse-grained models that discard the finest atomistic details and keep the most salient features of molecular structure, is particularly important. These models allow considerable economy in computational resources and provide a link connecting the world of atoms with the mesoscopic level, thus adding one more step to the ladder of multiscale simulations. It is crucial to find ways to compensate for the loss of information due to coarse-graining, in order to secure the predictive capability and reliability of the force-field parameters, in terms of computed macroscopic properties.
Traditionally, the development of new force-fields has been a time-consuming process involving trial-and-error computations. In our group, we are working in the direction of combining various approaches in an original context that will allow the fast generation of novel reliable models for arbitrarily complex systems selected by the user. At the atomistic level we can have a "library" of transferable bonded parameters based on *ab initio* computations or known empirical data, to describe the molecule's geometry and rotovibration modes. Single molecule geometry optimisation will fit classical potential energy functions to such data. Its iterative application, combined with normal mode analysis and other mathematical techniques can map the detailed atomistic description to united-atom and more coarse-grained models.
The non-bonded parameters, on the other hand, describe intermolecular interactions, incorporate the many body effects of condensed phases and account for all combinations of compounds and the resultant behavior of multicomponent mixtures and coexisting phases. Getting these parameters right is extremely important in the context of practical applications of the chemical and materials industry. Our approach benefits from a methodology that has been recently developed at Imperial College London, and uses SAFT, a molecular based Equation of States with a sound statistical-mechanical background, to match the Helmholtz free energy of a system either to existing thermodynamic data or simply another more detailed model, so as to give a reliable coarse-grained description.

6. CUDA Accelerated Molecular Simulations
Progress in computational techniques and increasing needs from both academic and industrial partners makes imperative to find ways to accelerate computations. Moving towards more and more complex problems leads often to very lengthy computations. This problem was till now met with the introduction of computational clusters and parallel MPI protocols. Recent advances in graphic cards as co-processors led to an interesting alternative known as the GPU computing paradigm, with CUDA and OpenCL programming languages being its most notable implementations. This also requires an extensive rewrite of existing code and application suites to meet the needs of researchers who would like to have computationally intensive jobs running on laptops and other 'lightweight' media.

In our group, we are continuously updating our applications in the other areas described above, to be compatible with new CUDA-enabled packages, such as the HOOMD Molecular Dynamics program that we have used extensively in recent studies of ours; we are also looking at the development of CUDA-enabled versions of the most demanding parts of our programs as well as a prospective MD application that would be integrated with the final version of our unified modeling platform.

7. Study of Phase Equilibria:
Calculation of thermodynamic properties is possible at the macroscopic level without using molecular models directly. Correlations of empirical data can be combined with the theory of chemical thermodynamics to yield useful predictions. This is a widespread method for the study of vapour-liquid equilibria of hydrocarbon and other mixtures, which is a pivotal topic in the chemical industry – all chemical engineering students have heard of the DePriester charts that provide values of so-called K-factors in graphical form. In our group we are developing applications to carry out phase equilibria calculations for a series of systems of interest to industrial applications. As an example, algebraic correlations, such as the McWilliams/DePriester equation and its more recent extensions and improvements, are used to calculate K-factors; subsequently, the phase equilibria problem is solved in numerically to compute the composition of the liquid or gas phase at given conditions or vice-versa. At a later stage, we are willing to combine these applications with our molecular modeling tools by using their predictions to compensate for missing experimental or correlated data.