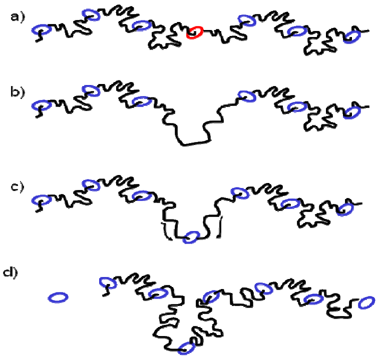
RESEARCH INTERESTS

The overall objective of my research is to develop mesoscopic models which link structure and dynamics, in order to design soft matter systems (at the molecular level) with desired macroscopic properties. At present my research interests focus on different main areas:

Modelling the linear and non-linear rheology of complex architecture macromolecular melts: I have developed **a general model for describing the linear rheology of increasingly complex model macromolecular architectures** (from linear[1] to star-like[2], pom-pom[3], dendritics[4], combs[5,6] and polydisperse blends[7]) **towards randomly branched polydisperse polymers**. This model is based on the tube concept, which allows a mesoscopic description of the chain while based on a small number of physically significant material parameters. Compared to other tube theories, I have proposed **some important modifications**. In particular, an approach based on a **time-marching algorithm**[2-4] has been used, i.e. in which the evolution of the relaxation mechanisms is determined step by step along the time axis, without the need for an analytical description. For hierarchical "branch on branch" structures, the motion of molecular segments localized between two branching points has been described carefully[4]. More importantly, **the model requires only 3 material parameters** (the plateau modulus, the Rouse time and the molecular weight of an entanglement segment), without the need to invoke additional "ad hoc" parameters, thus avoiding the related controversy[2,3]. The model is very versatile as it predicts the rheology of a range of model branched polymers with remarkable accuracy. We are now extending this model to **non-linear rheology**[8,9]. In particular, we have shown that accounting for the different modes of Rouse and their possibility to relax or not at a specific strain rate allows determining the stretch level of the chains once these ones reach their steady state[10]. The long term objective of this work is to better understand how stretch, strain hardening and spinnability of the material are linked.

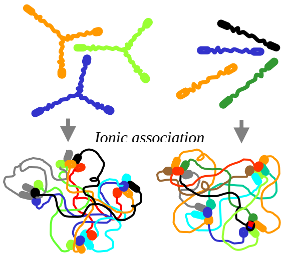
Fundamental questions about tube models: **Understanding the limits of the tube model** in order to extend its validity range represents, to me, an exciting objective. These last years, in collaboration with the Prof. H. Watanabe (Kyoto, Japan), I studied in details the relaxation mechanisms in a blend composed of short and long monodisperse linear chains, in different proportions[11,12]. Since the relaxation time of these two components have strongly separated, these blends represent one of the most challenging systems to describe. This work has allowed us to point out a new relaxation mechanism, which is based on the monomeric tension equilibration along a chain moving in a dilated tube. Based on this picture, we could explain why the apparent value of the dynamic tube dilution exponent, is larger than 1. We have now designed specific entangled polymer solutions in order to demonstrate, based on experimental data, that even in entangled solutions, this exponent is equal to 1[13]. The next step is obviously to test this approach on star/linear blends.



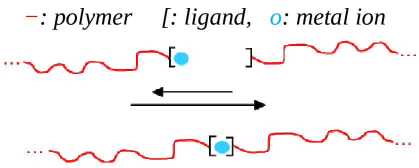
Tension equilibration: In addition to the short-long entanglements, long-long entanglements are also lost only due to the motions of the short chains

In collaboration with Prof. D. Vlassopoulos, I have studied very specific branched architectures in order to understand the influence of their branching point position[3,6,14]. In addition, by confronting theoretical predictions, experimental rheology and temperature-gradient interaction chromatography (TGIC) molecular weight distribution, we could discuss the limit of each technique as well as the influence of a small polydispersity (in weight and in architecture) of the sample[15]. The emerging idea is that fine details in the molecular structure may have an enormous impact on rheology. Even with top-of-the-line synthetic chemistry, small differences in molecular structure cannot be avoided, and the synergy of chemistry, rheology, interaction chromatography and modeling can address this important issue.

Statistical tool in order to relate synthesis recipe and chain architectures: For modeling the dynamics of polydisperse branched macromolecules, a detailed understanding of **their architecture** is essential. In this direction, I have developed **statistical approaches** which allow detailed architecture description, based on "average" information such as the branching density[7], average molecular weight[7] or the synthesis chemistry[16]. In particular, I used this research for the determining the composition evolution for branched polyamides (6 or 46) samples based on their synthesis recipe[17]. We are now extending this approach to statistically determine the localization of sticky groups along a supramolecular polymer chain in function of their density, and to determining their association state (free or associated) through time[18].



Multiscale dynamics of supramolecular polymeric assemblies: Today, with my group, my main research activity is related to characterizing, understanding and then, modeling the dynamics of associative entangled polymers able to create supramolecular assemblies. Our specific objective is to understand **the dynamics of supramolecular entangled polymers where both the topological constraints and association/dissociation dynamics of supramolecular junctions** play a role. In this direction, we have studied and modeled the viscoelastic properties of assemblies created from telechelic star and linear entangled polymer melts via ionic interactions[19], via metal-ligand interaction[20] or based on very weak, Crow-ether-like, interactions[21,22], with the objective of determining the self-assembling dynamics of supramolecular moieties in the environment of the polymer melt. Very recently, we extended our tube model to account for **supramolecular stickers along the linear chains**, these stickers being characterized by a very long relaxation time[18,23]. We are now developing a new approach for predicting the linear rheology of linear chains bearing shorter lifetime stickers[24]. With these systems, in a very natural way, we extended our research to the characterization of their **mechanical properties**. This has opened a new, fascinating door, which is to go all the way from the rheological properties of these materials towards their mechanical properties. This research is achieved within the framework of a European Marie Curie ITN (Supolen, FP7) that I am coordinating.



Rheology of soft colloidal systems: As a Marie Curie EIF researcher at IESL-FORTH (Crete, Greece), I have extended my research field to the structure and rheology of soft-colloids[25]. In particular, I have investigated the influence of several parameters (such as core-to-shell ratio, concentration, temperature, solvent quality) on the rheology of **stable block copolymer micelles**, pointing out very interesting rheological properties, such as a **non-monotonic behaviour of the zero-shear viscosity**. A key interest is to use a similar approach to study the combined polymeric and colloidal features in associating block copolymers. Ultimately I wish to develop quantitative criteria for designing soft colloidal systems and mixtures with tunable properties.

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