

PROGRAMME TITLE:

Experimental and Theoretical Design of Stimuli-Responsive Polymeric Materials

ACRONYM:

STIPOMAT

PRINCIPAL APPLICANTS:

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SUMMARY/ABSTRACT:

The aim of the proposed Programme is to combine the complementary expertises of leading European research groups in the experimental and theoretical study of complex structures on the basis of stimuli-responsive polymers and copolymers with linear or complex topology in order to gain a deeper understanding of two fundamental aspects of polymer science:

The first question is the understanding of the formation of such structures: self-assembly in stimuli-responsive block copolymers, dendrimers, branched polymers, thermoresponsive gels, interpolymer supramolecular complexes, networks with varying topologies and hybrids of responsive polymers with biopolymers.

The second question is the correlation between the behavior of these polymer systems under the change of external conditions (temperature, pressure, electric or magnetic field, shear, ionic strength, pH and composition of solution...) and the chemical structure of the constituents.

The research project will involve elaboration of new stimuli-responsive polymeric materials, application of advanced experimental methods in addition to atomistic and coarse-grained modelling, molecular mechanics, molecular dynamics and stochastic methods to determine structure and dynamical properties at different external conditions as well as kinetics of structure transformation of such systems under external stimuli. The groups involved have great experience in the synthesis, characterization, modelling and theory of complex polymer systems.

A previous successful multidisciplinary ESF SUPERNET Programme, in which some of the Steering Committee members already participated, was mainly focused on the bulk properties of polymers as construction materials. In the present project we suggest new approaches to polymer systems as multifunctional intelligent polymer devices for different applications (multifunctional polymer films, biocompatible surfaces, vehicles for drug delivery, intelligent materials for medical and structural applications, etc.).

The unique combination of synthesis, characterization, theory and modelling will allow a synergy for the elaboration and optimisation of novel multi-responsive materials with fine-tuned properties and for their future application in industry.

KEYWORDS:

polymer - smart materials – theory - simulation – synthesis – characterization –stimuli-responsive – complex structures

STATUS OF THE RELEVANT RESEARCH AND SCIENTIFIC CONTEXT:

In chemistry, the last few years have led to the development of new interdisciplinary branches combining and bridging different fields. The building principles and the perfect structures found in nature have served as motivation and as models. The combined efforts have led to compounds with ordered architectures. The design, synthesis, characterization and controlled self-organization of the required well-defined materials and systems on different length scales will be the key technology for the next decade. The results obtained up to now are just the very beginning in this development. The next years will be dedicated to further bridge the gap between synthetic and natural systems.

The thorough understanding of the self-organizing processes as well as the understanding and the precise control of structure and function on multiple length scales will be essential prerequisites for any significant progress. Complex polymer structures combining different polymeric blocks into various topologies are an important part of this effort.

The realization of the final structure uses contribution of different interactions: hydrogen-bonding, Van der Waals interactions, ionic interactions, metal-ligand interactions, hydrophobic interactions and entropic contributions. The chains and chain-segments gain, through these interactions, the possibility to recognize binding partners within and outside their own chains and to realize a final ordered structure. Such interactions are known to be sensitive to macroscopic boundary conditions (environmental changes) through the modulation of their strength and directionality. These systems can therefore serve as stimuli-responsive intelligent materials whose properties can be reversibly fine-tuned in the presence of the appropriate stimulus. This situation is reminiscent of biological systems in which such an approach is systematically used in order to infer a specific function to a structural unit.

Stimuli-responsive polymeric systems show a strong response to external stimuli (which result in forces on monomeric groups that normally are much weaker than typical intermolecular forces) only if there is a delicate balance between different types of interaction present in the system. It is clear that a search for suitable systems via suitable "chemical design" and appropriate sophisticated chemical synthesis techniques will greatly benefit from theoretical guidance coming from a theoretical analysis (by both analytical techniques and computer simulation) of suitable model systems.

At this point we illustrate this consideration by preliminary studies of lamellar ordering in thin films made from symmetric block copolymers via Monte Carlo simulations and self-consistent field type calculations [1-5]. These symmetric block copolymer melts order in the lamellar phase, characterized by a wavelength $l(T)$ which represents the size of the A-rich or B-rich domains in this microphase-separated structure. In a thin film geometry confined by surfaces, an energetic preference of the surfaces for one of the components (A or B) of the binary system will be normally observed. Because the lamellae tend to satisfy these surface interactions, an arrangement of the A-B interfaces parallel to the confining surfaces would be observed. However, there will be in general a misfit, since the film thickness D will not coincide with an integer multiple of the natural wavelength $l(T)$. As a consequence, an "elastic" distortion will occur (the periodic arrangement of the A-B interfaces is either compressed or expanded). In some cases [1,2], it will be even more favorable to arrange the domains oriented perpendicular to the surfaces. Phase transitions are therefore predicted where either the number n of parallel interfaces changes from n to $n+1$ or $n-1$, or transitions between parallel and perpendicular orientation occur.

These transitions were first predicted from simulations [1,2], then observed experimentally [6-10] and are a good example for a delicate balance between mostly enthalpic surface-forces and mostly entropic elastic forces in the mesophase structure of the thin film. In this case this balance is easily perturbed by external stimuli, such as electric fields. This leads indeed to the interesting possibility of switch from a parallel to a perpendicular pattern by a suitable variation of the electric field [10-12]. The technological interest for such switchable patterns and their use as templates for microelectronic applications is obvious. Of course, the stripe pattern of perpendicular lamellae in thin block copolymer films is only one out of

many block copolymer morphologies [13]. Other types of order-disorder transitions in surface-induced nanopatterns in diblock copolymer films [14] are easily manipulated by external stimuli. Moreover, not only electric fields are suitable as external stimuli: application of stress, in particular shear stress (a convenient tool to manipulate already the mesophase order of block copolymer melts in the bulk [13,15]), pressure [16], etc. A further variable at the disposal of the chemist is copolymer architecture: diblock vs. triblock vs. multiblock, grafted block copolymer vs. linear block copolymer, etc. As Bates and Fredrickson [17] put it, block copolymers are "designer soft materials", but theory and simulation are needed as a guide to understand how to control their properties.

As a second example where the interplay between theory and simulation on the one side with chemical synthesis and experimental characterization on the other side has helped to develop and understand a stimuli-responsive polymeric system, we mention mixed binary (AB) polymer brushes. If two polymers of type A and B that would be (at least partially) immiscible in the bulk are grafted on a substrate, the Flory-Huggins parameter causes a two-dimensional microphase separation, where several mesophases compete including ripple and dimple phases of square or hexagonal symmetry [18]. Which structure "wins" depends on a delicate balance of forces of both entropic and enthalpic origin, depending on volume fractions and chain lengths of the coatings with reversibly tunable properties [19,20,21]. In this last case, stimuli-responsive surfaces are formed: e.g. depending on the pH-value of the solution one may have one structure or another.

Both these examples show that for the development of "smart" functional polymeric materials the interplay between groups involved in theory and simulation on the one hand with the synthetic chemists and experimentalists on the other hand, was absolutely crucial for making progress.

Many more similar examples could be found, such as systems involving branched polymers, e.g. dendrimers, and polymer networks with various constituents.

OBJECTIVES AND ENVISAGED ACHIEVEMENTS:

In the present proposal, we would like to study stimuli-responsive complex polymer structures. Such polymers with new functionalities are expected to be applied for novel high technology applications, e.g. for a new class of sensors, and to mimic to some extent biological systems. The Programme will be based on an interdisciplinary approach merging together research teams involved in the synthesis of stimuli-responsive complex polymer structures, in the characterization of these systems on both the microscopic and macroscopic level and in theory and simulation. Such a unique combination of expertises is expected to lead to a dramatic increase in the understanding of complex structures.

The research project will involve the synthesis of stimuli-responsive polymers and their copolymers with linear or complex topologies. Polymerization techniques allowing a perfect control of the macromolecular parameters (molecular weight, stereo-regularity, location of functional groups...) and topology (block, dendritic and hyperbranched polymers as well as interpenetrating and semi-interpenetrating networks) are commonly used by several partners of the Programme. As examples, living anionic polymerisation techniques are golden tools to synthesize (co)polymers with perfectly defined macromolecular architectures and controlled molecular weights. These techniques have been implemented by a member of the Programme for the synthesis of ABCD-type block copolymers [22] or ABC-type star-shaped block copolymers in which the chemical nature of each arm is varied [23]. Other groups are specialized in controlled radical and living copolymerisation techniques and have a strong expertise in the synthesis of stimuli-responsive polymers [24,25]. Special emphasis will be devoted to stimuli-responsive water-soluble polymer blocks such as poly(2-vinylpyridine), poly(methacrylic acid), poly(ethylene oxide), poly(vinyl ethers), poly(*N*-alkylacrylamide), poly(*N*-vinyl caprolactam) and polypeptides that exhibit pH, ionic strength or temperature dependent behavior and that will be combined together or with other hydrophobic blocks. The topology of the resulting block copolymer (linear, comb-like, hyperbranched, star-like, networks) will be

systematically varied. Solid-phase synthesis will be used for the preparation of polypeptides that will be then coupled to well-defined synthetic polymers.

Other types of stimuli-responsive materials will be obtained by combining conducting polymers to interpenetrating network in order to build electrically-responsive actuators or artificial muscles [26]. Core-shell macromolecules originating from multifunctional macroinitiator and dendrimers will be also prepared and investigated. These synthetic strategies are already well implemented in several of the participating partners of the STIPOMAT Programme.

We are interested in studying the properties of single macromolecules like core-shell branched macromolecules and dendrimers under the action of external stimuli which can find application in drug delivery and other biomedical systems as well as in self-assembling of macromolecules into different supramolecular structures and responsive polymer films, polymer brushes, polymeric micelles and multifunctional or/and biocompatible surface. The self-assembly of these structures will be controlled by introducing supramolecular non-covalent interactions between specific polymer segments or blocks, resulting in complex polymeric superstructures. The primary structure and topology of the copolymer will determine the location of such non covalent interactions and hence the secondary (or even ternary) structure of the super-aggregates. The properties of the polymer blocks and segments as well as the supramolecular interactions will be then fine-tuned by the application of external stimuli.

Coil-globule or coil-helix transitions upon application of stimuli as it occurs in biological systems will be investigated for systems containing polymer blocks that could exhibit such transitions.

Supramolecular chemistry will be utilized to assemble different polymer blocks functionalised by suitable ligands. In this respect, one of the participating group has shown that terpyridine-ruthenium complexes are promising candidates for the formation of the so-called metallo-supramolecular copolymers. In this novel synthetic approach, a polymer A block is functionalised with terpyridine ligands whose position in the polymer chain is controlled (at one end, on both ends, along the backbone). *Mono*-terpyridine-ruthenium complexes can then be selectively formed by complexation of terpyridine with Ru(III). These mono-complexes are then further reacted with other terpyridine-functionalized polymer B blocks while Ru(III) is reduced to Ru(II). This results in *bis*-terpyridine-ruthenium complexes which act as non-covalent linkages between the polymer A and B blocks [27]. The resulting metallo-supramolecular A-[Ru]-B block copolymers are quite stable and do not show ligand exchange over very long time-scales (years), they can be manipulated as block copolymers and self-assembled [28]. Under specific conditions, the complexes can be however opened-up, which opens new avenues for the application of such polymeric materials [28]. Moreover, these "lego"-type clicking units can serve as building blocks towards complex copolymer topologies. These metallo-supramolecular copolymers will be investigated in the frame of the STIPOMAT Programme.

Advanced spectroscopic, scattering, microscopy and calorimetric methods are available within the network. Application of NMR relaxation methods, ultrasonic and dielectric relaxation methods, measurements of rheological and mechanical properties, mechanical spectroscopy, neutron, X-ray and laser light scattering analysis, scanning and transmission electron microscopy techniques for morphological characterization and scanning probe microscopies form the basis of the experimental work on complex polymer structures. The stimuli-responsive polymeric materials prepared above will be thoroughly characterized by using the pertinent techniques.

The experience of all participating groups in the preparation and characterization as well as in the theoretical description of polymer systems will be combined to develop novel approaches for the optimised synthesis of stimuli-responsive complex polymer structures. An integrated approach will be used in which characterization, theory and simulation efforts will be devoted to selected stimuli-responsive complex polymer systems. The feedback from theory and simulation will be then used in order to optimise the design of the complex polymer structures. An essential aspect of the theoretical modelling required

for these types of materials is the multiscale character of the problem. An atomistic description must include input from quantum chemistry (for the description of intra-chain potentials) and Car-Parrinello-type "ab initio molecular dynamics" methods, for instance. A good example for such an approach is the recent work on the adsorption of bisphenol-A-polycarbonate on nickel surfaces [29,30]. By carefully designed mapping procedures, the very detailed small-scale information is "translated" into effective potentials, useful for coarse-grained models of the bead-spring type [32,32]. The groups proposed for participation in the STIPOMAT network bring in excellence and experience for both the special multiscale coarse-grained techniques [30,31,32] and the more standard classical Molecular Dynamics and Monte Carlo simulations [33]. More detailed ab initio simulation on the base of Density functional theory, DFT, will be also applied.

A particularly intriguing problem occurs for water-soluble polymers (e.g. biopolymers and synthetic polyelectrolytes) where protonated water cluster $H^+ \cdot (H_2O)_n$ play a role. In this case, the formation and breaking of the network of hydrogen bonds has to be taken into accounts. Encouragingly, problems of this type have recently become accessible to the computational modeling by clever combinations of the "ab initio" Car-Parrinello Molecular Dynamics (CPMD) technique with the Path Integral Molecular Dynamics (PIMD) approach (see [34] for a thorough introduction into all these techniques). It will be an interesting challenge to combine all these expertises in order to explore possibilities for a more accurate description of forces in such systems containing smart polymers.

Furthermore, we emphasize that on the mesoscale it is still necessary to complement the already rather coarse Monte Carlo techniques [33], DPD and Lattice Boltzman dynamics by still simpler methods suitable for faster computations on still larger length scales, such as the numerical self-consistent field approach [3,4,18] and its extension to deal with dynamical properties [35,36]. The STIPOMAT network would provide a perfect framework for the various theoretical groups specialized in all these techniques to interact among themselves, share the techniques and develop them further, and make them fruitful for various specific systems and experimental applications, helping the synthetic chemists and experimental physicists to develop new types of stimuli-responsive polymeric systems and to understand their behavior and properties.

As previously stated, an integrated approach between synthesis, characterization, modelling and theory of stimuli-responsive polymeric materials will be developed within the STIPOMAT Programme. A major concern in the Programme will be the training of young researchers, who will benefit of the fruitful exchanges of knowledge and expertise occurring between the participating groups. In order to illustrate this integrated approach, some examples of research topics that will be investigated within the STIPOMAT Programme are described below.

1) Linear stimuli-responsive polymers and proteins.

Different responsive polymers on the base of either *N*-isopropylacrylamide or *N*-vinyl caprolactam will be studied and simulated under different conditions. Computer simulation of polypeptides and proteins in native and unfolded state will be also carried out and compared with behavior of stimuli-responsive "protein-like" synthetic polymers. The characteristics of unfolded stimuli-responsive polymers and proteins (average end-to-end distance, gyration radius, correlation of orientations of bonds along backbone, as well as different distribution and autocorrelation functions) will be simulated and compared with that for synthetic polymers and with predictions of theoretical models. The behavior of such polymers in external fields (which also can be referred to as an external stimulus) will be simulated and compared to experimental data such as the well-known mechanical unfolding extension of proteins in atomic force microscope (AFM) experiments. It was shown that these experiments can give some details of folding pathways and are sensitive to local mutations. At the same time the mechanical unfolding by pair of forces impose rather strong restriction on movements which can occur during unfolding. Unravelling of protein occur in this case from protein ends as it was shown by steering molecular dynamics simulation by Shulten et al. [37] and Karplus et al. [38]. It can not be the case for unfolding under other "more natural" conditions, like pH, temperature or pressure jump, for example. Direct simulation of whole pathways of such "natural" unfolding on computer is

not possible due to large time scale of these processes. It is not also quite clear whether pathways of “natural” and forced unfolding are the same or not. Due to this reason it is important to check other possible forced unfolding procedures for example unfolding by different flow fields. We plan to perform both real and computer experiments of such unfolding in the framework of the present project and perform theoretical description of these processes.

2) *Stimuli-responsive block copolymers.*

Grafted, linear and hyperbranched block copolymers containing stimuli-responsive (e.g. poly(*N*-isopropylamide), PNIPAM, and poly(ethylene glycol), PEG) and other other blocks, will be prepared and simulated. Living and controlled polymerization procedures will be used in order to obtain macromolecules with well-defined molecular characteristic features (low polydispersity and controlled molecular weight of the constituent blocks). Special attention will be paid to copolymers consisting of hydrophobic and hydrophilic components as well as copolymers possessing charged groups. Various functional groups will be introduced in these copolymers in order to promote additional non-covalent interactions (ionic interactions, VDW forces, hydrogen-bonding) and additional levels of structural complexity resulting from the emergence of ternary (or even quaternary) superstructures. Metallo-supramolecular copolymers will also be considered. Very complex block copolymers of the ABCD type will be investigated. The self-assembly of these complex polymers will be experimentally investigated and simulated. How these complex polymeric structures depend on the chemical structure and the external conditions will be studied by using Brownian, molecular dynamics and more coarse grained methods. The results will be compared both with experimental data and with predictions of analytical theory. Multi-responsive systems are expected to be formed from such polymeric materials. Block-copolymers of proteins and synthetic polymers will be also prepared. Pegylation (i.e. preparation of copolymer of proteins and PEG) is designed to increase protein solubility and stability and reduce protein immunogenicity. Such PEGylated proteins as PEG-adenosine deaminase, PEG-L-asparaginase, PEG- α -interferon-2b and others are widely used in drug delivery. The conformational and dynamical properties of such and other block copolymers will be simulated to predict their behavior during drug delivery.

The self-assembly of the stimuli-responsive block copolymers in micellar structures will be investigated in selective solvents for some of the constituent blocks. Some of the partners of the project have previously shown that such stimuli-responsive micellar objects are very promising candidates for various applications ranging from drug release, templating nanomaterials, pigment stabilization, etc. [24,25,39]. Further efforts in the preparation of these polymeric materials will be devoted in the frame of the present Programme. Advanced characterization techniques will be used to visualize how the stimuli-responsive properties are actually acting within these materials.

3) *Stimuli-responsive dendrimers.*

Functional dendritic polymers are used for different applications. The controlled shape, size and differentiated functionality, ability to form isotropic and anisotropic assemblies, complexes with many other types of nanometer scale building blocks, their potential for self-assembly, capacity to form surfaces and interfaces, ability to either encapsulate or to be engineered into unimolecular devices make dendrimers uniquely versatile among existing nanometer-scale building blocks. Computer simulation of dendrimers taking into account electrostatic and hydrophobic-hydrophilic interactions will be performed. The correlation function manifested in different experiments (mechanical and dielectric relaxation, NMR, dynamic Kerr-effect and dynamic X-ray and neutron scattering) will be calculated and compared with the experimental data and with theory predictions. Dendrimers with terminal responsive azobenzene groups, with terminally attached PNIPAM linear chain or other responsive groups and polymer will be prepared. Such hybrids can have very unusual properties. We plan to study the conformational and dynamical properties of such hybrids at different external conditions.

Complexes and conjugates of dendrimer with low-molecular drugs will be constructed and simulated. All stages of drug delivery (encapsulation of drug into complexes, behavior of

complexes in bloodstream, during adsorption on cell surface and controlled release from complexes under different external conditions) will be studied. Multiple attachment of recognition groups to the dendrimer surface can help to increase binding (multifunctional binding) of dendrimer to specific receptors of cancer and other targeted cells. Stimuli-responsive dendrimers or hybrids of dendrimers with stimuli-responsive polymers can be used for better controlled release. We also would like to realize the very promising idea about creation of multifunctional dendritic nano-devices for targeted drug delivery on the base of differently functionalized dendrimers. Two approaches are possible: 1) inhomogeneous functionalization of one dendrimer by different functional groups (solubilizing, targeting, drug, radioactive, imaging, reporting and other) or 2) homogeneous functionalization of each dendrimer by one type of functional groups with successive combination of these dendrimers in larger supermolecule (tecto-dendrimer, cylindrical dendrimer or dendrimer-polymer hybrids). This last variant allows to create in the future a library of such dendrimers which can be quickly combined with each other into different specific supermolecules for the targeting of different diseases. In the present project different approaches to the creation of such nano-devices will be implemented and their properties will be studied.

The interaction between dendrimers possessing charged functional groups and oppositely charged linear chains will be studied. The formation and stability of such complexes depend on the ionic strength and pH of the medium. Our investigations will address the manner in which a polyelectrolyte adsorbs onto a dendrimer and the extent to which the polyelectrolyte chain penetrates into the interior of a dendrimer as a function of quantities such as charge density and distribution, polyelectrolyte length and flexibility, number of dendrimer generations, pH and others. The main system to study will be the complexes of dendrimers with different polyanions. Such complexes in salt free solution have been studied using experimentally. The unexpected findings revealed that the flexible polyanions were able to penetrate into the interior of dendrimers of at least generation five. At the same time less flexible DNA was observed to bind to the dendrimer outer shell only rather than penetrating into its interior. The structure, conformation properties and dynamics of such complexes under different conditions will be studied.

The adsorption of dendrimers and responsive polymers containing different functional groups on surfaces of different functionalities will be also studied. Such stimuli responsive polymer brushes or "dendrimer forests" with high concentration of functional groups can be used for selective adsorption of different components under different external conditions or for creation of functional polymer surfaces.

4. Stimuli-responsive ion-containing polymeric materials.

Electrostatic interactions are widely used to modulate the structure and properties of polymeric materials. The modulation of the dielectric constant allows the fine-tuning of the magnitude of these interactions. Although these interactions are studied for many years in polymeric materials, important questions are still pending. Moreover, ions can be introduced into polymeric materials through ionizable weak basic or acid groups. The ionization degree of these polymers is depending on the environment (pH of the surrounding). Thus, stimuli-responsive polymeric materials can be obtained. In the STIPOMAT proposal, stimuli-responsive (co)polymers with various architectures will be synthesized. The effect of ionic interactions in these materials will be characterized from both the structural (SAXS, SANS on bulk or micellar samples) and the ultimate properties (mechanical and dielectric spectroscopies) point of view. The formation of interpolyelectrolyte complexes will be used to control the self assembly of mixtures of mutually interacting copolymers. Theory and simulation of complex polymer system with electrostatic interactions (mono and multi-component polymer gels, polymer monolayers, complexes of oppositely charged polymers) will be carried out. Crosslinked polymers such as gels or rubbers are the stabilizing structure in many materials and biological systems. The detailed understanding of their properties on the molecular level is highly desirable. At the present great progress has been achieved in the theoretical understanding of structure and properties of polymer systems with rather complex topology formed by neutral polymer chains. In the present project the main attention will be devoted to

simulation of such systems consisting of stimuli-responsive polyelectrolyte chains. Electrostatic interactions occurring in metallo-supramolecular copolymers, that contain charged complexes and their associated counter-anions will be also characterized. Preliminary results have shown that these secondary ionic interactions had a profound effect on both the self-organization of the copolymers and on the mechanical properties. Surface anchored water-soluble and polyelectrolyte polymer brushes are other examples. Long-ranged forces between surfaces in a liquid control effects from colloid stability to biolubrication, and can be modified either by steric factors due to flexible polymers, or by surface charge effects. In particular, neutral polymer 'brushes' may lead to a massive reduction in sliding friction between the surfaces to which they are attached, whereas hydrated ions can act as extremely efficient lubricants between sliding charged surfaces. It has been shown that brushes of charged polymers (polyelectrolytes) attached to surfaces rubbing across an aqueous medium result in superior lubrication compared to other polymeric surfactants [40]. This was attributed to the exceptional resistance to mutual interpenetration displayed by the compressed, counterion-swollen brushes, together with the fluidity of the hydration layers surrounding the charged, rubbing polymer segments. These findings may have implications for biolubrication effects, which are important in the design of lubricated surfaces in artificial implants, and in understanding frictional processes in biological systems. The effect of the length of the polyelectrolyte segments, their charge density as well as the ionic strength on these remarkable lubrication properties will be investigated into details in the present Programme. Interdisciplinary study of all these systems including their synthesis, experimental and theoretical study and computer simulation is presumed in the project.

EXPECTED BENEFIT FROM EUROPEAN COLLABORATION IN THIS AREA:

The goal of the Programme will be to establish European leadership in the elaboration and the application of stimuli-responsive polymers via joint multidisciplinary investigations. The topic of the proposed Programme is a key issue for polymer science in the next future. Stimuli-responsive "smart" polymers are indeed the subject of intense research in both academic and industrial world. It is now clear than polymers can be considered not only as structural materials but can also have a specific function that can be triggered in the presence of an appropriate environmental change (stimulus). Elaborated responsive behavior can be obtained by mimicking biological processes in hybrid or fully synthetic complex polymer structures. This proposal is clearly multidisciplinary and is merging together people from many EU countries and associated states which have been carefully selected on the basis of their integration in the project. Intense exchange of knowledge and expertise is expected to occur within the networking activities (meetings and exchange of students). Moreover, the unique combination of synthesis, advanced characterization, theory and modelling is certainly one of the most interesting characteristic feature of the present proposal, that will stimulate exchange of knowledge and expertise among the participants to the Programme.

EUROPEAN CONTEXT: R&D COLLABORATION WHICH ALREADY EXIST IN THE ACTUAL FIELD:

The majority of the participating groups have a long-standing collaboration in the frame of the successful ESF Programme SUPERNET in the field of polymer synthesis, characterization and theory of multicomponent polymer systems. As a result of this collaboration new multicomponent systems were synthesized and studied, a significant number of joined publication have appeared. There are also joint contributions of participants to international conferences. Some of the participants took part in special joint European grants (INTAS). Other participants are taking part in another joint proposal (ESF SIMU) to establish European networks on molecular simulation. However, the present project will combine participants with wider complementary experiences than earlier. Therefore it should allow for a more complex approach to the systems under investigation.

PROPOSED ACTIVITIES:

Workshops and conferences: The Steering Committee organises international workshops with around 40 participants which are held about once a year. The main contributors to the Programme will discuss the advancement of common research during this annual meeting. In addition larger international conferences with around 100 participants will be organized every two years and will cover a broader range of people.

Between the conferences and workshops the supervision will be organized by members of Steering Committee. Connection between different teams both on national and international level will be organized using electronics telecommunication including organization of teleconferences. The role of Internet and especially Grid in communication, sharing the data and integrating of joint work in framework of this Programme will increase with time.

Short-term fellowships: These are mainly for young scientists, who need further training and expertise in new experimental and modelling methods for a fruitful continuation and broadening of their research scopes. Short term fellowships are intended to facilitate the transfer of knowledge and techniques relevant to research from one laboratory to another within Europe (at least one contributing country should be involved). The grants are for periods up to six months.

Short scientific visit grants: These cover the costs of short visits of senior researchers working in the area of the Programme, in order to carry out joint work primarily in one of the STIPOMAT participating laboratories.

DURATION: 4 years

BUDGET ESTIMATE:

| | Year 1 | Year 2 | Year 3 | Year 4 | Total |
|--|-----------|-----------|-----------|-----------|------------------|
| Exchange of researchers (short visits and exchange grants) | 77 500 € | 77 500 € | 77 500 € | 77 500 € | 310 000 € |
| Workshops and conferences | 50 000 € | 50 000 € | 50 000 € | 50 000 € | 200 000 € |
| Total | 127 500 € | 127 500 € | 127 500 € | 127 500 € | 510 000 € |

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1 April 1972 – 31 March 1973: IBM postdoctoral fellow at IBM Zürich Research Laboratory

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April 1974 – Sept. 1974: Research consultant at Bell Laboratories in Murray Hill

1 Oct. 1974 – 30. Sept. 1977: Professor of Theoretical Physics University of Saarbrücken, Germany

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5 June 1978, 30 April 1981: Birth of my son Martin, Birth of my son Stefan

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1986 – 1992: Member of the Technology Advisory Board of Rhineland-Palatinate

May 1986 – Jan. 1996: Chairman of the Materials Research Center (MWFZ) Mainz

July 1987 – Dec. 2001: Speaker for DFG Special Research Program SFB 262 on “The glass state and glass transition of non-metallic amorphous materials”

July 1987 – July 1995: “Scientific Advisory Board” at HLRZ Jülich, Germany

June 1989 till present: External Member of the Max Planck Society

May 1992 to present: Corresponding Member of the Austrian Acad. of Sci., Vienna

24 March 1993: Max Planck Medal of German Physical Society (DPG)

1999 – 2002: IUPAP C3 Commission (Chair), IUPAP Executive Council Member

2001: Distinguished as “Highly Cited Researcher” by ISI, Philadelphia

2001 (Sept. 6th): Berni J. Adler CECAM Prize (for Computational Physics)

2003 (Jan. 15th): Elected for 2 years as the Chairman of the Physics Department

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- Binder K. Phase transitions of polymer blends and block copolymer melts in thin films. *Adv. Polym. Sci.* 1999, 138: 1-89

- Milchev A, Yamakov V, Binder K. Escape transition of a Polymer chain: Phenomenological theory and Monte Carlo simulations. *PCCP* 1999, 1: 2083-2091

- Binder K, Baschnagel J, Paul W. Glass transition of polymer melts: test of theoretical concepts by computer simulation. *Progr. Polym. Sci.* 2003, 28: 115-172

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- Varnik F, Binder K. Shear viscosity of a supercooled polymer melt via non-equilibrium MD simulations. *J. Chem. Phys.* 2002, 117: 6336-6349

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- Ricciardi R, Gaillet C, Ducouret G, Lafuma F, and Lauprêtre F. Investigation of the relationships between the chain organization and rheological properties of atactic poly(vinyl alcohol) hydrogels. *Polymer* 2003, 44: 3375-3380
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- Beaume F, Lauprêtre F, and Monnerie L. Secondary transitions of aryl-aliphatic polyamides II. High-resolution solid-state C-13 NMR investigation. *Polymer* 2000, 41, 2989-2998

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1989: First Laureate of the Belgian Chemistry Olympiads

1994-2000: appointed as teaching assistant at the University of Liège

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10/2002-10/2003: Invited Professor at the Université catholique de Louvain (Belgium).

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- Gohy J-F, Lohmeijer BGG, Varshney SK, Décamps B, Leroy E, Boileau S, Schubert US. Stimuli-responsive aqueous micelles from an ABC metallo-supramolecular triblock copolymer. *Macromolecules* 2002, 35: 9748-9755.
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