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Light-sensitive polyaminoacids as bio-inspired molecular materials

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Dedicated to Professor Osvaldo Pieroni.

Abstract

In this paper, we consider light-sensitive polyaminoacids and polypeptides, i.e. polyaminoacids and polypeptides modified by luminescent, photochromic and other fragments. Polyaminoacids are objects that mimic natural proteins and can change their conformation under different outer factors, which result in remarkable changes in the physical, chemical, and mechanical properties of macromolecules. Incorporation of various dyes, photochromic fragments and metal-containing compounds into polypeptides can lead to formation of unique supramolecular assemblies similar to biological systems and design of novel molecular materials with nanoscale function.

Keywords: Polypeptides; Polyaminoacids; Light-sensitive; Molecular materials; Organic–inorganic hybrids

1. Introduction

Light-sensitive natural proteins are known to act as photosensors and energy converters [1–2]. Bacteriorhodopsin and bacterial photosynthetic reactive center are the most investigated objects. Absorption of light by dye fragments gives rise to photo-induced electron transfer that is well modeled in terms of donor–acceptor interactions [3].

Polyaminoacids are unique objects that mimic natural proteins and can change their conformation even under weak outer factors. The latter result in remarkable changes in the physical, chemical, and mechanical properties of macromolecules. In other words, polyaminoacids are adaptive or intelligent systems [4]. Ever increasing interest of researchers to nanoscale supramolecular complexes and assemblies has led in recent years to synthesis of numerous substituted (modified) polyaminoacids and polypeptides for use in molecular electronic devices (MED). Main research interest is given to photochromic and light-sensitive polypeptides, which is evidenced a number of reviews by Yamamoto [5], Pieroni and Popova [6–8], Cooper et al. [9,10], Wegner [11], and Willner and Rubin [12].

In this paper, we will consider the perspectives for use of light-sensitive polyaminoacids and polypeptides as intelligent molecular materials in various devices.

2. Chemical modification and assemblies of modified polyaminoacids and polypeptides

Aminoacids, especially tri-functional ones, can be readily modified by various reactive groups (a) in a side chain, (b) in order to obtain the so-called ‘tail-head’ structures, and (c) to form cyclic systems, Fig. 1. To date, the above modifications have been applied to a huge number of polyaminoacids (PAA) and polypeptides (PP). Additionally, PAA covalently bound with other chemical compounds can form the supramolecular assemblies and phases.

The synthesis of modified peptide oligomers, supramolecular phases–films (membranes), liquid-crystal structures, and related gels was initiated in the end of the 1970s in Japan and then continued in Germany, Italy and USA. The synthesized modified PAA/PP can be divided into several groups: (1) chromophore-containing PAA, (2) linked with surfactants (laurylsulfate, sodium tridecylphenylsulfate, sodium dodecylsulfate, etc.), (3) linked with polymerizable fragments (acetylene-, vinyl-, acryl-, and styrylpeptides), (4) block-copolymers of aminoacids (with polysaccharides, polyethylene, hydroxypropyl cellulose, etc.) and dendrimers, and (5) complexes with metals and inorganic

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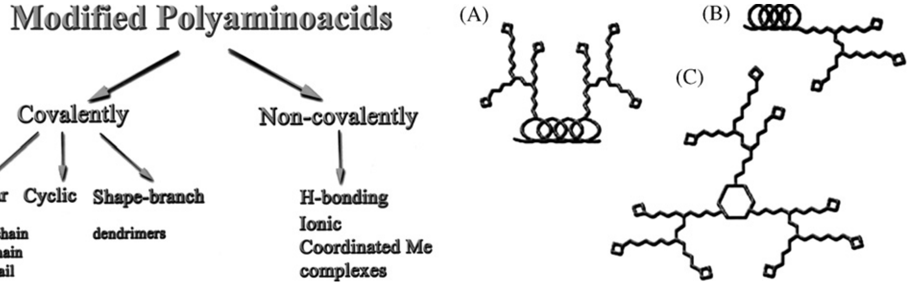


Fig. 1. Different ways of polyaminoacid modification.

ligands. Most promising turned out compounds of groups (1), (4), and (5) and also their combinations resulting in architectures exhibiting unexpectedly new properties.

Most widespread synthetic techniques include (i) *N*-carboxyanhydride synthesis, for preparation of compounds (1), (2), (4); (ii) the method of activated esters and *N*-hydroxysuccinimide synthesis, for compounds (1), (2), (4); (iii) carbodiimide synthesis and the method of activated esters for polymer-like transformation of side groups, for compounds (1)–(3), and (5). For condensation of non-aminoacids with peptides [yielding compounds (3)–(5)], the common methods of organic and polymer synthesis may turn out applicable.

Modern synthetic approaches to above compounds are based not only on the knowledge of chemical behavior of combinatorial units (building blocks) but also on preliminary quantum-chemical modeling of resulting structures and optimization of their electronic structure. A strategy for targeted synthesis of modified PAA/PP can be represented by the following scheme:

Combinatorial synthesis of building blocks → *Computer modeling of intermediate and end products, their electronic properties, and molecular architecture prediction* → *Synthesis of target compounds and their identification* → *Self-organization and formation of supramolecular phases* → *Investigation of function on a nanoscale level* (*molecular materials*) → *Stimuli responsive properties* (*intelligent molecular materials*).

Due to the presence of multicenters of non-covalent bonding, PAA/PP exhibit the ability of self-organization, thus producing supramolecular assemblies of varied architecture: rods, tapes, ribbons, barrels, bracelets, and fibrils [13–16]. In turn, these assemblies can form mono- and multilayers, liquid crystals, gels, etc. [17,18]. In all cases, most typical is the so-called hierarchical alignment of assemblies, depending on the level of their organization [19,20].

In this communication, we consider modified PAA/PP suitable for preparation of supramolecular phases, some of their properties and parameters with special emphasis on their potential use as molecular materials in micro- and nanoelectronics, biomedicine, ecology, and other related areas of science and technology.

3. Properties and potential application of light-sensitive molecular materials

Over the past years, chips functioning on a molecular and nanoscale level (molecular electronic) have been being designed. Such chips can be based on either bioorganic (various proteins) and synthetic compounds or inorganic compounds (such as new semiconductors). The process for deposition of conducting template on the surface of synthetic analog of polylysine has been elaborated in the 1980s [21]. Nowadays, a progress in R & D on biomolecular chips has led to the design of BioMEMS (bioelectronic mechanical systems). A key point of these studies is the synthesis of compounds suitable for use in the technology of Langmuir–Blodgett (LB) films, self-assembling monolayers (SAM), and monocrystals. The use of LB films for recording optical data was overviewed in [22–24]. Much attention is being paid to the photochromism of J-aggregates in multilayer structures (reversible data recording) and to interactions at the interface (nanophotonics) [25,26].

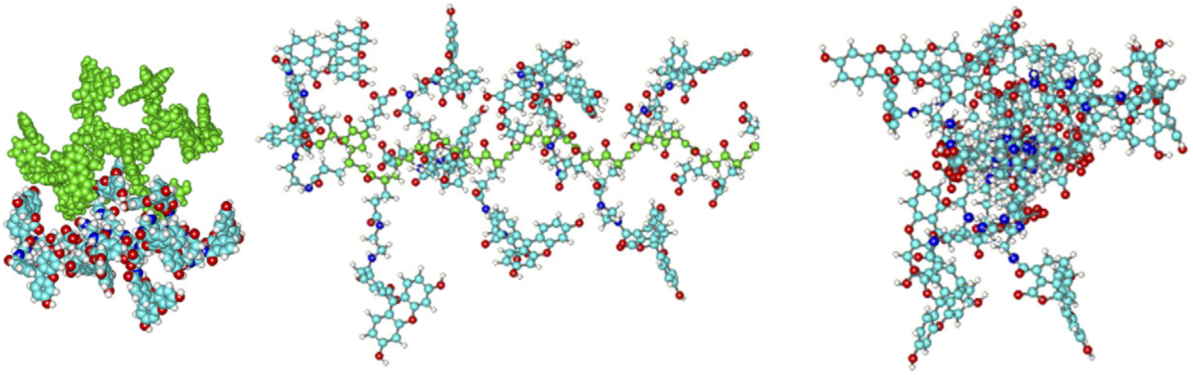


Fig. 2. Interaction of two fluorescein-containing PAA helices (molecular mechanics simulation).

Further progress in submicron technology of chips is impossible without the development of high-resolution resist materials, including those based on monomolecular multilayer films. Preparation of self-assembling monolayers (SAM) has demonstrated good perspectives of this line of research and development.

3.1. Photoactive films and membranes

The photochromic behavior (including conformational transitions with summarization) of spiropyranes and azo derivatives of PAA/PP was first investigated by Pieroni and coworkers, Menzel, and Yamamoto et al. [27–32]. Later, some interesting photochromic systems have been synthesized and characterized by Russian researchers [33].

Data on the photosensitivity of monolayers formed at air/water interface can be expected to elucidate photo-induced transformations of individual molecules present in the system. Such data can also provide a basis for the design of nanostructures and turn out helpful for deep insight in the mechanism of action for biological photoreceptors. It seems interesting to pay attention to reversible photo-induced changes in surface tension.

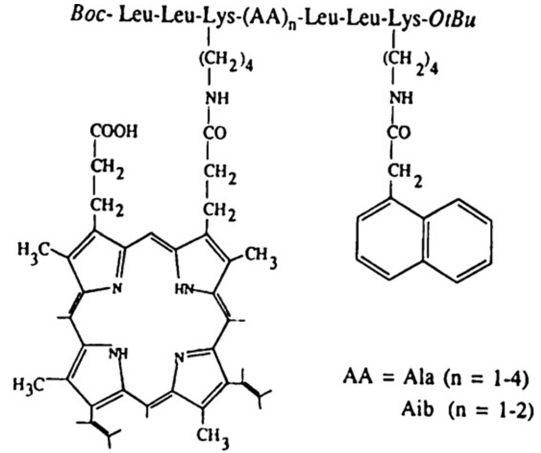
Behavior and properties of modified polyglutamic acid at different irradiation times were investigated by Menzel et al. [30,31,34]. The photo-induced and thermal isomerization of azobenzene fragments were explored by circular dichroism (CD) absorption spectroscopy. The *cis*–trans-isomerization of these fragments was found to occur upon irradiation at 370 nm. According to CD data, the amount of α-helices is dependent on the number of azobenzene moieties in PAA molecules and independent of their configuration.

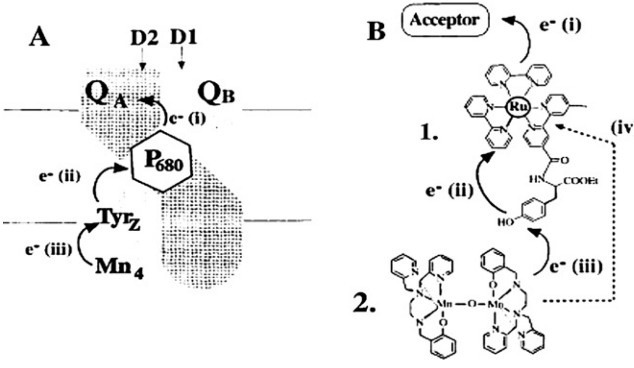
Yamamoto synthesized poly-L-ornithine linked to azobenzene fragments by carbodiimide method [35]. The photochemical behavior of azobenzo-PP was studied by circular dichroism (CD) and absorption spectroscopy [36]. Azobenzene derivatives containing alkoxy groups with different kind of crosslinks with poly-γ-benzyl-L-glutamate were found to have a cholesteric liquid-crystal structure. Dye molecules readily link with polyaminoacid and undergo easy hydrolytic elimination. The orientation of dye molecules can be fixed when the latter ones are covalently bound with PP. The films of poly-γ-methyl glutamate (PMG) cast from 1,2-dichlorethane were found [37] to be coloured in the visible due to the presence of the cholesteric (liquid-crystal) phase. These films exhibited the selective reflection of circularly polarized light, which gives rise to absorption at 450–600 nm in the CD spectra.

The aggregation of polyglutamic acids (PGA) with acridine orange, pianocyanol, and toluidine blue was explored in [38–41]. The dynamic piezoelectric measurements with oriented films were carried out at *f* = 10 Hz [42]. Light-sensitive oligoaminoacids modified with carbocyanine dyes have been synthesized and characterized aiming at their potentials use in positive photoresist films and thermochromic and electrochromic films [43–45]. Modification of side chains in PAA with chiral moieties afforded the preparation of super-thin membranes suitable for separation of enantiomers and its controlling by varying the folding of PP chains [46,47].

3.2. Charge transfer complexes and luminescent derivatives

For studies on photo-induced energy transfer, the PP-containing various electron-accepting and electron-donating chromophores in different sequence and orientation have been synthesized [48]. Very interesting results have been obtained by Fox and coworkers [49] for the migration of excited state energy along the α-helical PAA chain and for detailed study of energy transfer along macromolecule.





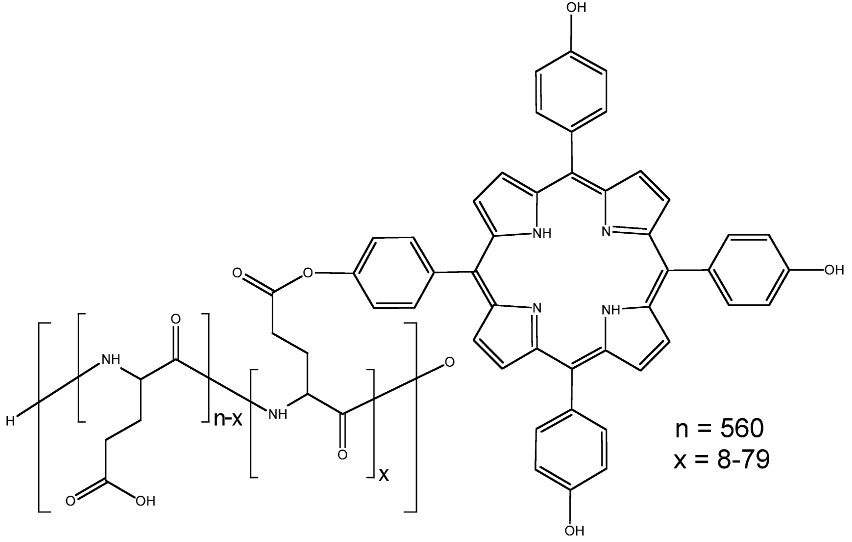
For photosynthetic reaction centre study a non-helical triad based on ruthenium/bipiridine complex was obtained [50]. The photoconductivity of poly-γ-(β-*N*-carbazolylethyl)-L-glutamate and its charge transfer (CT) complexes with 2,4,7-trinitrofluorenone in a polymer film cast on a gold electrode was measured. The glutamate complex with 2,4,7-trinitrofluorenone was found to form only within the near-surface layer of films [51].

The attachment of Rhodamine B and Rhodamine 6Y to terminal groups of poly-γ-benzyl-L-glutamate – through reaction between amino groups in PAA and Rhodamine B 5-isothiocyanate in chloroform–dichloroacetic acid mixtures – was investigated in Refs. [52,53].

Polyglutamic acid regularly substituted in side chains by luminophores—fluorescein and/or rhodamine was obtained and studied for pH changes [54,55]. Modified PAA can form aggregates comprising two helices that are able to self-assemble into a hierarchical structure of up to 29 macromolecules in solution and LB film for fluorescein-containing PAA, Fig. 2. Reversible self-organization–reorganization processes occur under pH ≥ 8 and luminescent properties depend upon pH that can be explained by different forms (bonds) of fluorescein fragments by rearrangement of molecular aggregates.

The energy transfer and luminescence of poly-γ-1-naphthylmethyl-L- and -DL-glutamates in various co-polymers with γ-L-naphthylmethyl-L-glutamate and γ-benzyl-L-glutamate were investigated in [56]. The luminescence spectra exhibited the bands of normal and excimer emission. With increasing number of naphthyl groups in a copolymer, the intensity of excimer fluorescence increases. In has been inferred that the excimers are formed both in the ground and excited state, while γ-benzyl-L-glutamate units act as barriers for the intramolecular transfer of singlet excitation over copolymer molecules and ultra-thin films.

The reaction of poly-L-glutamic acid with 5,10,15,20-*tetrakis*-4-hydroxyphenyl-porphyrin in the presence of dicyclohexylcarbodiimide (DCHC) was used to obtain poly-L-glutamic acids with different amount of 4-(10,15,20-*tris*-4-hydroxyphenyl-5-porphyrinyl)phenoxy groups [57]. The conformations of synthesized compounds were investigated by circular dichroism (CD). With increasing number of porphyrin fragments in poly-L-glutamic acid, the formation of α-helical conformation becomes impeded, and the films cast from different solvents become unstable. The structure of above-mentioned polymers is interesting not only for luminescent liquid crystals but also for ion-conducting assemblies creation.



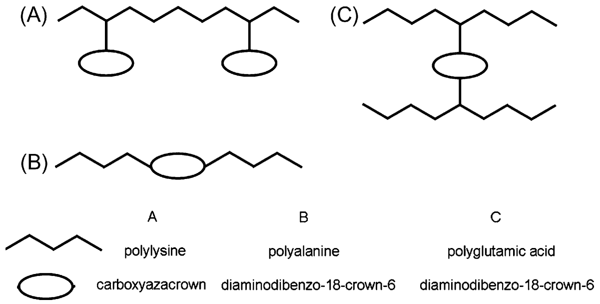


Fig. 3. Polyaminoacids containing crown ether fragments in side chains (A), in crosslinks (B) and in backbone (C).

3.3. Systems with ionic and electronic conductivity

Non-covalent binding of metal-containing compounds to PP structures can be expected to provide the properties that are inherent to living organisms but are hard to achieve in synthetic organic systems, viz. the ionic or electronic conductivity accompanied by a change in absorption (colour) or emission (luminescence). The attempts were made to prepare superconducting systems upon intercalation of TaS_2 with polyglycine layers [58] and also semiconducting and light-sensitive PP with metal-induced conformational transformations [59]. Some studies have been devoted to ionic transfer through lipid monolayers [60] and to design of peptide channels for ionic conductivity [61,62]. The ionic conductivity can also be ensured upon addition of macrocycles or some dyes (largely in the form of salts) to biopolymers. Some promising results have been obtained by Russian co-workers. They synthesized crown-containing polyaminoacids with different design, on retention of sorption properties of crown ethers, both in solutions and thin films, Fig. 3 that can lead to ionic conductivity [63]. These co-workers found the electric conductivity (longitudinal and cross) of cabrocyanine-containing PAA [45,64,65] as well. The data of quantum-chemical calculations for polylysine ($n = 12, 25$ mol.% carbocyanine) suggest that the ionic channels can be formed due to a specific arrangement of dye moieties in a sector of 90° relative to the helical axis [66]. These data can be expected to give rise to new studies on design of electroconducting bio-inspired systems. Multilayer structures bearing opposite electric charge can be used to form sandwich-like polyelectrolyte systems for preparation of biomimetic materials imitating natural leather, threads, etc. [67]. The results published by Yamamoto and coworkers [68,69] show the possibility of making threads of PAA linked to chitosan (via complex formation). This was done by using the technology of nano-capsulation that is being actively developed during the past years [70].

3.4. Rotaxanes and dendrimers

PAA/PP derivatives are also being used as parts of 'molecular machines' in the design of rotaxanes [71–73], photo-switches

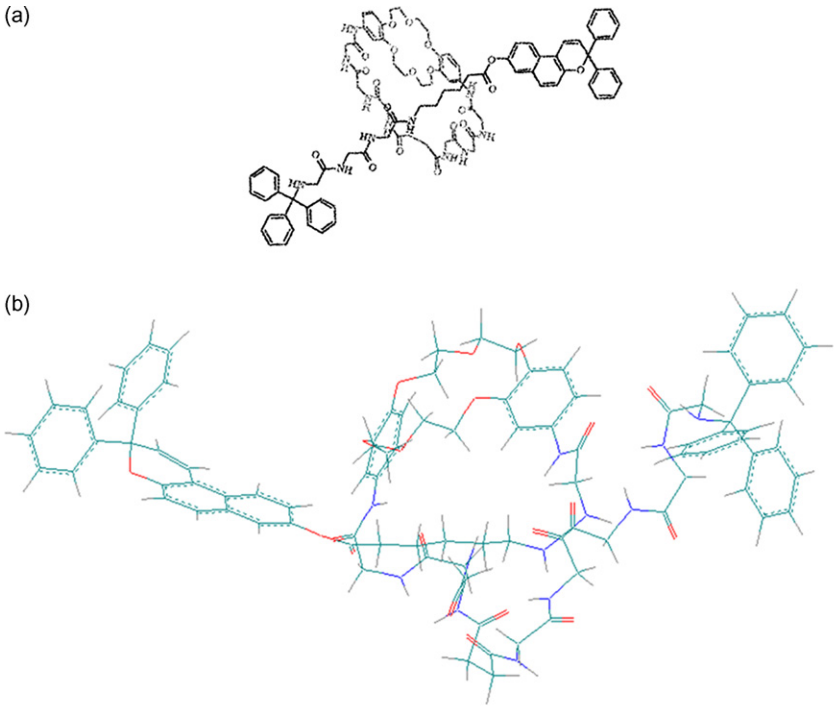


Fig. 4. Photocontrolled ion-containing rotaxane: (a) chemical structure and (b) optimized model.

[74], and non-linear optical systems [75]. Peptide rotaxanes first time synthesized by Leih and coworkers [71] are based on the dependence of hydrogen bonding on the type of solvent. Some rotaxanes were labeled with fluorescent moieties [76]. Psuedo-roaxane-containing 24-crown-8 has been synthesized in characterized [77] under temperature changes.

Rotaxane structure based on the cyclopeptide/crown ether system and co-polypeptide with polyester linked with spiropyran or spironaphthoxazine as a stopper was synthesized; this structure can act as ion-containing photo-switch [78], Fig. 4.

Unique properties of dendritic complexes gave rise to synthesis of PAA/PP dendrimers [79,80]. Japanese scientists described

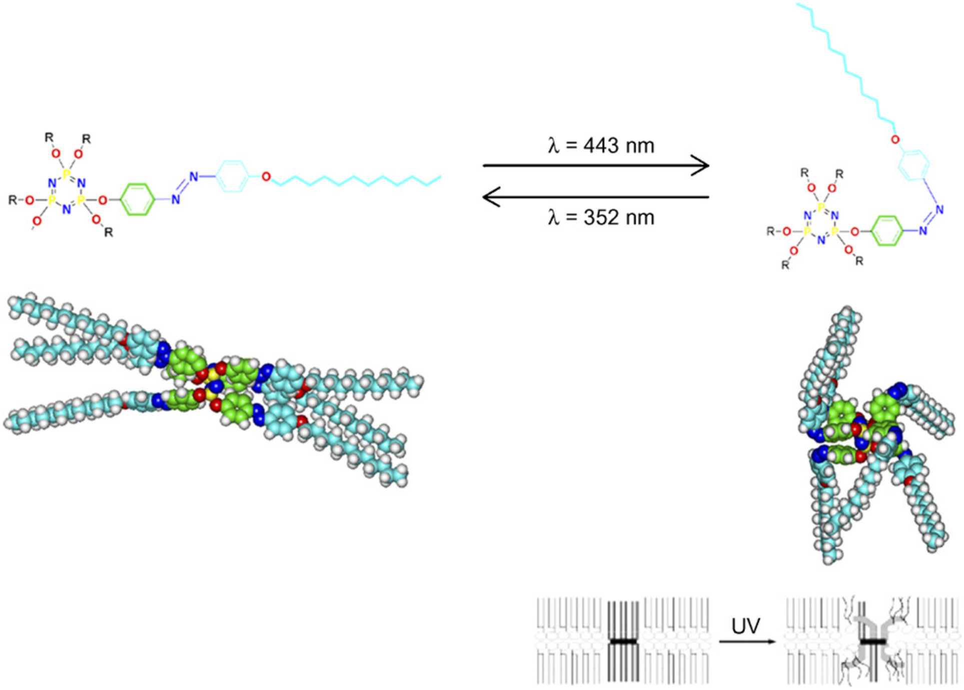
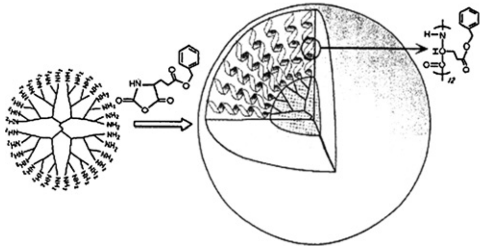
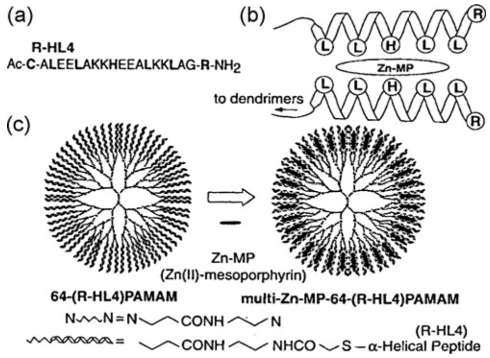


Fig. 5. Conformation of hexakis(p-(p'-dodecyloxyphenylazo)phenoxy) cyclotriphosphazene by molecular mechanics calculations. Proposed model of destruction of highly ordered structures under UV-irradiation.

dendrimers containing peripheral α-helical fragments of polyglutamic acid and polypeptides [81].



Of special interest is the PP dendrimer with containing Zn-containing porphyrins embedded into interchain voids [82].



Such structures with a developed charged surface may be applied in bio-diagnostics and as dot elements in biomolecular electronics.

3.5. Organic–inorganic hybrids

The presence of organic and inorganic fragments in the same molecule can give rise to synergistic effect and lead to unconventional supramolecular architecture. Independently, Japanese and Russian researchers proposed unique star-like structures where six polyaminoacid chains are attached to cyclotriphosphazene (CTP) core [83–90].

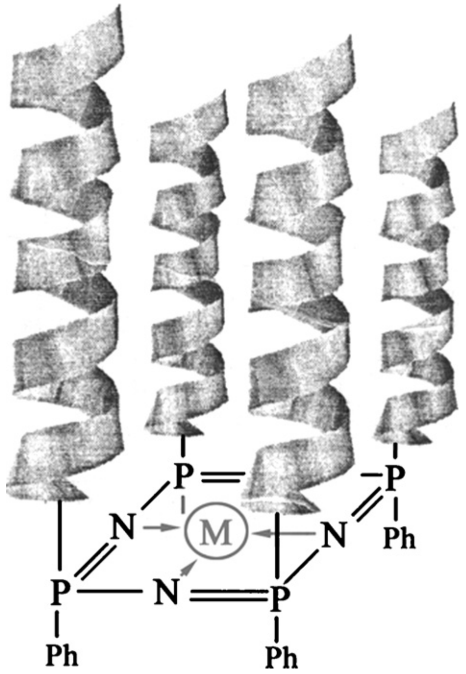


Fig. 6. 4-Helix bundle formation induced by cyclotetraphosphazene/metal ion complex templates.

FTIR, CD and SWAXS data point towards highly ordered structure of organic–inorganic hybrid where polyglutamic acid chains self-assemble into three α-helical bundles in trans-configuration to central CTP core. These compounds may form mesophases and multilayers with LC domains [89,90].

Later, the joint Russian–Japanese research group carried out the template synthesis of PAA/CTP hybrids. This synthetic

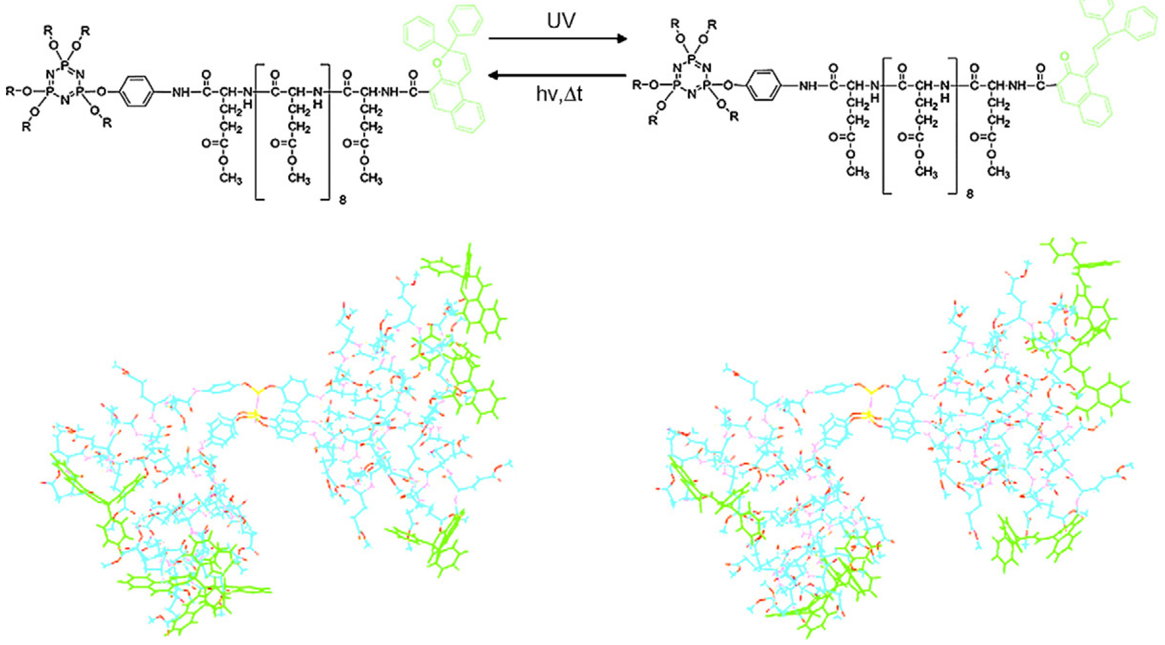


Fig. 7. Polyaminoacid/cyclotriphosphazene hybrid with peripheral photochromes—computer modeling by molecular mechanics simulation.

approach may be used for preparation of luminescent and photochromic cyclophosphazene–PAA hybrids capable of forming films and gels [86–88]. The presence of azobenzene moieties allows tuning the gel colour upon UV–vis irradiation [86], Fig. 5. A multifunctional system based on cyclotetraphosphazene is described in [90], Fig. 6. The disordered chains organize into helices only if metal ion forms complex with cyclotetraphosphazene. Interesting results were obtained by computer modeling of multicentered photoactive CTP/PAA hybrids, Fig. 7. Investigations aimed at structure prediction and computer modeling of multicentered CTP/PAA hybrids are being carried out by Russian scientists [91,92].

3.6. Bioactive ingredients

As already mentioned, PAA/PP-based bioactive systems are of considerable interest for potential use in biomolecular electronics and biomedicine. In the former case, it concerns the development of enzyme electrodes and immuno- or DNA-based sensors [93] while in the latter one, means of bio-diagnostics and drug-delivery systems [94,95]. Electrochemical reduction on the gold surface (including modification of PGA) was used to explore the orientational interaction between microdipoles on the electrode [96]. The 'substrate–synthetic PP receptors' interactions are being in focus as key stages of molecular recognition of antibodies in organism [97]. In this context, the biomimetic PP/lipid monolayer systems [98], the cholesteric order of rod-like PAA [99], etc. are investigated. Multifunctional nanoscale organic–inorganic hybrids are proposed as non-toxic drug-delivery systems and as nano-biocatalysts, e.g. for the cleavage of yeasts and fats [100]. Besides, CTP/PAA hybrids can be used as universal building blocks for nanoreactors creations.

4. Conclusions

Due to numerous technologically useful properties, the light-sensitive polypeptides seem to be promising materials for potential use in the future. Incorporation of various dyes, photochromic fragments and metal-containing compounds into polypeptides can lead to formation of unique supramolecular assemblies similar to biological systems and design of novel intelligent molecular materials with nanoscale function.

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