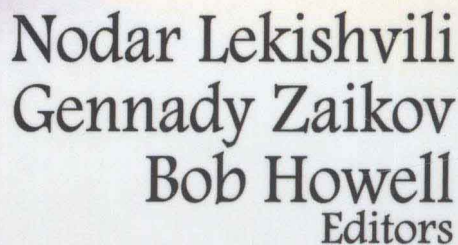


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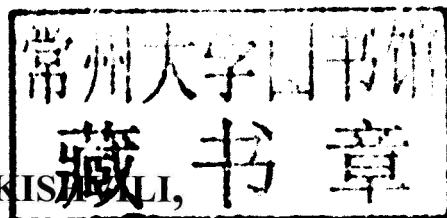
ENVIRONMENTAL SCIENCE, ENGINEERING AND TECHNOLOGY

**ADVANCED BIOLOGICALLY ACTIVE
POLYFUNCTIONAL COMPOUNDS AND
COMPOSITES: HEALTH, CULTURAL
HERITAGE AND ENVIRONMENTAL
PROTECTION**

**NODAR LEKISHVILI,
GENNADY ZAIKOV**

AND

BOB HOWELL
EDITORS



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PREFACE

During the last decade, researchers working within the field of biologically active compounds were attracted to finding new compounds, materials and methods that could be used toward the protection of human health and environment, along with the preservation of cultural heritage from various microorganisms, viruses and fungi. The actuality of this problem was stressed at the 41st congress of IUPAC devoted to these and related topics. Many actual aspects on creation of new compounds were discussed, materials and methods capable of more effective solving of the aforementioned problems. The topics that are discussed in this book encompass novel bioactive systems for human and environmental protection and preservation of cultural heritage from the microorganisms' and fungi attacks. Also presented are the results of theoretical and experimental investigations conducted by the experienced scientists from different countries, along with the mechanistic comprehension of their performance under various experimental conditions.

Chapter 1 - Chemically activated 1-hydroxycyclohexyl hydroperoxide decomposition in the presence of ammonium salts is proposed to proceed through the complexation stage. Complex structure and reactivity have been investigated by molecular modelling methods. Kinetics of the chemically activated hydroperoxide decomposition in the presence of quaternary ammonium salts (Et_4NBr , Pr_4NBr , Bu_4NBr , and Hex_4NBr) has been studied. The correlation between reactivity and structural characteristics of ammonium cations was found.

Chapter 2 - Electrical phenomena and cytoplasm movement on an example of seaweed *Characea* have been briefly described.

The results concerning cyclosis nascent mechanism in the cells of seaweed *Characea* and connection of cytoplasm movement velocity (CMV) with the difference of electrical potentials (DEP) of plasmatic membrane have been adduced. For specification of the appropriateness of interconnection of the cyclosis and DEP, the authors carried out the simultaneous measurement. A suggestion was made for a model in the frame of which it is possible to set up, in every separate case, a way to realize the observed effects by over regulation of the membrane potentials, i.e., finally by changing of ion penetrability of the membrane and in what—due to presence of other influences (for example, by influence on the difference intercellular processes).

For ascertainment of the possibility of the presence on the plasmatic membrane of vegetable cell structures, similar to animal receptors and the interaction character of them, effectors, a detailed research had been conducted of acetylcholine acting and some biogenic amines on the velocity of cytoplasm and DEP of cell of seaweed *Characea*. *Nitella flexilis*.

Results show that a reaction of the cell on adrenomimetics, active in respect to α and β -receptors, is discerned by sign. Investigation of combined action of adrenomimetics and α -type blockaders (nor adrenalin-fenitron) and β -type (isadrin – dihydroergotoxin) showed the strongly pronounced antagonistic effect, realizing by concrete mechanism. There were adduced also other proofs allowing for the existence of interacting centers in the cells with molecules of catecholamines, similar to α and β -receptors of animal cells; while this resemblance is spread as on the structure organization, so it is on some functional features.

Data about the presence of cAMF, cGMF, enzymes of their synthesis and catabolism (adenilatcyclase, gaunilatcyclase, phosphodietherase) and protein-target – proteincinases, also G-protein, allowed authors of the work to make the conclusion about the specific interaction of acetylcholine and biogenic amines with the specialized structures of plasmatic membrane of vegetable cells and signal transmitting by trigger mechanisms on the G-protein with following participation of secondary intermediary messengers.

Following from the above-stated consideration, the authors proposed the presumptive conclusions:

There is revealed the phenomenon of receptor regulation of intercellular physiologic processes of vegetable cells.

In the experiments with biogenic amines was stated the appropriateness in the direct correlation between alteration of CMV and DEP.

The cells of *Nitellaceae* can be used in the model experiments for testing of biological active substances, inasmuch as there was revealed by them the property to react by constellation receptor system on the exogenous influence of low concentration of testing compounds.

All those presuppositions show that in the plant cells is the presence of contractive proteins in view of actinomyozin complexes, otherwise who may answer the question which is open till today: What gives the first push to the cytoplasm particles to move constantly along the perimeter of the vegetable cell?

Chapter 3 - This study investigates the actions of caffeine analogues and antagonists to the main Ca^{2+} -depo in the rabbit skeletal muscle - sarcoplasmic reticulum. The efficiency of Ca^{2+} -accumulation by vesicles of heavy fragmented sarcoplasmic reticulum was greatly changed by caffeine analogues and antagonist: cordiamin, camphor, bemegrid, 8-methylcaffeine, teophylline, theobromine, midocalm. All tested substances have some similar characteristics at its molecular structure. The carbonyl oxygen and methyl groups are of a great importance for “caffeine effect” of these substances.

Chapter 4 - New biologically active adamantane-containing anilides and nitroanilides have been synthesized and studied. By the semiempirical quantum-chemical method, AM1 effective charges on the atoms, bonds lengths and valence angles, enthalpies of formation of initial compounds and probable obtained products of the reaction of nitration of the anilides have been calculated. Based on quantum-chemical calculations and experimental data, the direction of the reaction of nitration has been established. To model physical properties of biologically active 16 anilides and nitroanilides, the authors studied quantitative “structure-properties” relationships (QSPR) on the basis of the experimental data. The authors used several sets of molecular descriptors. The dataset outliers were identified by using Principal Component Analysis (PCA). As a modeling technique, the authors applied Projections to Latent Structures (PLS). To ascertain the quality of models, the authors used cross-validation.

Based on the author's research, one concludes that the best models were acquired by the use of GETAWAY.

The anthelmintic activity and activity towards various microorganisms of the obtained compounds have been studied. Based on preliminary investigations, it was established that the obtained compounds may be recommended as modifiers of anthelmintic preparations—phenacetyne, trinoine, diamphenetide, raphoxanide—also as a bioactive component for preparation: a) materials with antimycotic properties for prophylaxis and treatment of mycosis and dermatomycosis; b) protective covers stable to biocorrosion from action of some mycopathogenic microorganisms.

Chapter 5 - This work deals with the problem of modeling of a possible mechanism of point mutations of DNA under the influence of Ni^{2+} ions. Two feasible schemes of interaction of Ni^{2+} with nucleosides are considered. The first scheme presents the formation of the planar complex of Ni^{2+} -G-C, and the second shows the incorporation of Ni^{2+} between the two neighboring complementing pairs. The authors used the MOPAC package to compare the force constants and energies of intramolecular hydrogen bonds in the complexes and corresponding values in a free G-C pair. The comparison enabled us to make a conclusion implying that the formation of the Ni^{2+} -G-C complex is accompanied by the weakening of a hydrogen bond, nearest the joining point of Ni^{2+} .

The incorporation of Ni^{2+} between the two neighboring complementing pairs of G-C causes the weakening of all the three pairs of hydrogen bonds, but to a lesser extent. It has been demonstrated that in the first case the probability of point mutations, the replacement of G-C by A-T increases, and the probability of divisions, the fallout of triplets of type GGX or XGG increases in another case.

Chapter 6 - The influence of hydrolysis and centrifugation processes of soybean semifat flour on various indices of the lipid component and dynamics of changes in the composition and characteristics in hydrolyzates within three months of storage were studied. It was shown that processes of hydrolysis and centrifugation, and also storage, cause reliable changes of the physical and chemical characteristics and lipid composition in hydrolyzates.

Chapter 7 - Some patterns of the relationship of the interaction between aromatic amines (o-, m- and p toluidin; o-, m- and p-amino phenol; o-, m- and p-amino benzoic acid) and aldoses (D-glucose, D-galactose, D-mannose, L-rhamnose, D-xylose, L-arabinose, D-maltose, D-lactose) in the Maillard reaction are investigated. In the Maillard reaction, the reactivity of aniline, toluidines and amino phenols increases and the reactivity of amino benzoic acids decreases, with increase of pH of the reaction medium; in comparison with aldohexoses, aldopentoses participate in melanoidin formation more actively.

Chapter 8 - The influence of serpisten and inokosterone on the phospholipids composition in liver and blood erythrocytes, intensity of lipid peroxidation in tissues (liver, spleen, blood plasma), catalase activity in liver and general peroxidase activity of white outbreed mice has been studied. A biological activity of ecdysteroid-containing compounds is shown to be associated with an influence on the parameters of the physicochemical regulatory system of lipid peroxidation (LPO). Possessing pronounced membrane-tropic properties due to alterations in the exchange of predominantly choline-containing fractions of phospholipids, ecdysteroid-containing preparations are capable of modifying a cell membrane phase state. A substantial dependence of a biological effect of the compounds on a dose, duration of their

application as well as on an intensity of the LPO processes in the tissues and an animal's sex require a more detailed research on the properties of the given ecdysteroids.

Chapter 9 - Nanoscopic electrochemical devices composed of SAM-modified electrodes and redox probes (RP) of different kind (complex ions, proteins, organic molecules, etc.), were proven to be suitable systems for studying intrinsic electron transfer (ET) mechanisms and interplay between them. In the present review the authors consider the author's recent results on the mechanistic studies of Au/SAM/RP nanoscale devices in which the RP were either redox-active protein cytochrome *C* (CytC) dissolved in aqueous solution, or the complex compound ferrocene $[\text{Fe}(\text{Cp})_2]^{0/+}$, Cp=cyclopentadienyl) dissolved in a room temperature ionic liquid (RTIL), [bmim][NTf₂]. The SAM composition was either [-S-(CH₂)_{*n*}-OH] with *n*=2,3,4,6 and 11, or [-S-(CH₂)_{*n*}-CH₃] with *n*=1,2,3,5,7,11 and 17, respectively. The modern electrochemical methodology including fast scan cyclic voltammetry and data processing techniques were applied to extract ET rate constants and an impact of the variation of ET distance, viscous additives (or temperature) and high pressure was determined, allowing for a rigorous discrimination of intrinsic ET mechanisms. In particular, at short electrode-reactant separations, *n*=2-3 for Au/SAM/CytC and *n*=1-3, for Au/SAM/ $[\text{Fe}(\text{Cp})_2]^{0/+}$, the adiabatic mechanism of ET controlled by the viscosity-related relaxational modes of the RP environment, found to be operating. At larger electrode-reactant separations, *n*=6-11 for Au/SAM/CytC and *n*=7-11 for Au/SAM/ $[\text{Fe}(\text{Cp})_2]^{0/+}$, the non-adiabatic ET mechanism can be observed manifested through the exponential decay of rate constant with the increase of *n*. At *n*=4 and *n*=5, the intermediate (mixture) regime of ET can be detected. Furthermore, for the case of *n*=17 for Au/SAM/ $[\text{Fe}(\text{Cp})_2]^{0/+}$ in [bmim][NTf₂], dynamical arrest (broken ergodicity) for ET has been demonstrated. In overall, together with other matching proceedings, the reviewed results, despite of essentially diverse nature of complex environments invoked within two different series of congruent systems (protein/aqueous solution versus RTIL) demonstrated common general patterns of the mechanism changeover, in a nice agreement with the predictions of a generalized theory of ET.

Chapter 10 - The spherical PLGA nanoparticles (NP) calorimetric investigation is presented in this paper. Such nanoparticles is used for biological active substances (drugs) encapsulating inside of them with the purpose of medicine transferring into the cell. It is clear that without determination of particle stability it is impossible their practical usage. From calorimetric study of PLGA nanoparticles with PLA/PGA ratio 70:30 it was determined the entirety conditions of such particles and the temperature interval, where the particle destructions take place. It was unambiguously shown that for noncoated PLGA NP and for chitosancoated PLGA NP the stability temperature are equal to 37°C and less than physiological temperature, which exclude their practical application. Also it was determined that hermiticity destroy temperature depends on heating rate. At the same time it was established that strongly alkaline and acid area (pH2 – pH9) do not destroy noncoated PLGA NP and chitosancoated PLGA NP what gives possibility for their using orally.

Chapter 11 - Three industrial samples of Poly(methyl methacrylate) (PMMA), prepared under different conditions, have been extensively analyzed by means of ¹H-NMR spectroscopy. Starting from the *mm*, *rr* and *mr* triad contents, as given by the spectra, the type of tacticity statistics distribution has been deduced. Sample X appears to be completely Bernoullian, while samples Y and Z deviate somewhat from this behaviour

exhibiting a tiny trend towards Markovian statistics. The fraction of *mrmr* and *rrrm* pentads and that of pure heterotactic and atactic triad moieties has been calculated by assuming either a Markovian statistics for samples Y and Z or a Bernoullian statistics for all the samples. On the other hand, the fraction of the same pentads has been determined by deconvoluting the overall triad signals of the spectra into the corresponding pentad signals. An appreciably good agreement with the values obtained assuming Bernoullian statistics for all the samples appears evident. As a result, the evolution of every pentad content from sample X to Sample Z could be stated. Thus the samples prove to be appropriate models to study the relationship between any physical property and the stereomicrostructure of PMMA as was done previously for Poly(vinyl- chloride) (PVC) and Polypropylene (PP).

Chapter 12 - It was investigated the fermentative stationary kinetics of hydrogen peroxide decomposition under the action of catalase in the presence of bioSAS. It were obtained the kinetic parameters of this process. It was shown, that the bioSAS have influence on the fermentative process, which can be explained by the change of the fermentative center activity or by the change of substrate concentration. It was determined that the temperature of a process has an insignificant influence on the value of kinetic parameters.

Chapter 13 - The results of studying the axial coordination of tetraphenylporphyrin complexes of high-charged metal cations (AcO)CrTPP, O=Mo(OH)TPP and O=W(OH)TPP with molecular ligands (hydrogen sulfide, imidazole and pyridine) in toluene are discussed. The thermodynamic and the kinetic characteristics of reactions between metalloporphyrin and molecular ligand were obtained by the method of spectrophotometric titration and chemical kinetics. Correlations between the molecular ligand basicity and the molecular complex stability are discussed.

Chapter 14 - The catalytic rate constants for the process in the presence of bioSAS by different concentrations have been obtained. It was shown, that the constants some increase at bioSAS concentration increasing up to their micelle-formation beginning. The temperature has a slight influence on the value of catalysis constants, that can be explained by practically zero activation energies and depend on activation entropy.

Chapter 15 – The authors studied dynamic trend of changes in the activity of creatine kinase, aldolase and succinate dehydrogenase in brain cells under 30-day long stress induced by isolation and violated diurnal cycle. It was shown that these enzymes heterogeneously responded to 30-day long stress. Particular sensitivity was recorded by succinate dehydrogenase that showed the decline of activity in various sections of the brain at 60-80% on average. Unlike succinate dehydrogenase, aldolase activity increased on the 10th day of stress and then declined. Similar results were seen in relation to phosphokinase activity.

It was observed that the change in the activity of the enzymes in question was accompanied by quantitative changes in nitric oxide-NO. In accordance with experimental data the authors suggested that the main signal molecule causing changes in enzyme activity should be NO.

Chapter 16 - Hydrazides are characterized with different pharmacological activity, amongst, compounds with inhibitory action of hydrophobia virus and human immunodeficiency virus. Therefore, the authors considered as perspective synthesis of adamantane containing hydrazides and anaemia opposite ferroocenes admixtures. Ferrocene-A(ferrocenyl-1-phenyl-dioxy-1,4-butan-2) has significant antitumour and antibacterial properties. The derivatives of adamantane have broad pharmacological activity, low toxicity,

high membranotropic and antibacterial properties. The search new biologically active compounds, the authors have synthesized ferrocene- and adamantane-containing derivatives.

Chapter 17 - Perspectives in the field of creation of highly effective anticancerogenic preparations have been evaluated. For their creation is offered a new regio-selective method of glycosylation of alkylurea in conditions of nucleophilic catalysis with some following nitrosing of glycosyl carbamides of the D- and L-rows.

This method opens principally new possibilities for modification of compounds by means of glycosylamides bond leading to preparations possessing small toxicity and high selectivity.

Chapter 18 - Physical and biochemical functionalisation of bast fibres are ways to improve thermo- and moisture regulation, anti-bacterial anti-allergies, hygiene, creating “smart” textile. Enhancing natural properties of vegetable fibres is an intermediary step in the obtaining of new products with special applications. The vegetable fibers are biodegradable, can be recycled, and in natural state are highly polar and hydrophilic. To improve the properties of the cellulosic fibers, the chemical and/or physico – chemical modifications were applied. The surface esterification of the natural polymer with acids can be carried out to obtain biodegradable materials, novel fibres with tailored functionalities for special applications.

In this paper, starting from Spanish broom (*Spartium junceum*, syn. *Genista juncea*) fibers, under action of cold plasma discharges, and using different kinds of carboxylic acids, cellulose esters with short and long side chains have been synthesized.

The new grafted polymers were characterized by FT – IR spectroscopy (ATR), XPS and SEM in order to assess the existence of incorporated functional groups. The thermal characterization of the obtained fibres reveals their particular behaviour.

Chapter 19 - Miniaturisation of technical systems creates the need for today’s science and engineering to assess the mechanical properties of small volumes of material. A specific feature of the structure and the combination of the desirable properties across several different length scales are fascinating by the many examples in biology. Determining the extraordinary properties of natural materials at the nanometer scale is regarded as a very attractive target for materials science. Mechanical behavior of various biological materials such as insect and plant cuticles was studied by applying experimental approaches of material science in order to explain their structural design and working principles. Experiments were performed on the head articulation cuticle of the beetle designed for friction minimisation and on the wax covered plant surfaces adapted for attachment prevention. Both insect and plant cuticles are multifunctional composite materials and have a multilayered structure. *Gula* cuticle of the beetle *Pachnoda marginata* is a part of the head articulation, which is a micromechanical system similar to a ball bearing. The surfaces in this system operate in contact and must be optimised against wear and friction and provide high mobility within the joint. The measurements on the *gula* cuticle were performed in order to understand structure and mechanical behavior of the material working for friction minimizing. The wax layer on the plant surfaces is a barrier for the attachment system of insects. Antiattachment function could be caused by contamination of attachment pads of insect with the wax crystalloids. Increase in roughness due to location of the wax crystals on the plant surface causes decrease in the real contact area between the plant surface and attachment pads of the insect. These are two of the hypotheses why insects cannot walk on the plant surfaces structured with wax. Nano-indentations on different plant surfaces were performed in order to understand the deformation behavior of the wax layer. This study is believed to be one of the first for

mechanically testing insect cuticle and the very first for wax-coated plant surfaces in native condition.

Chapter 20 - New carbofunctional oligosiloxanes containing trifluorinepropyl and methacrylic groups at silicon atoms have been synthesized and studied. Biological active nitroanilides with spatial adamantane-containing groups and cadmium complex compounds based on them have been obtained. By using the data of IR and NMR spectral analyses the composition and the structure of synthesized compounds have been established. New composite materials of multifunctional application for individual and environmental protection, based on the obtained silicon-organic carbofunctional oligomers and complex compounds, have been created. It was shown that the created composites could be used as: a) protective covers (film materials and impregnating compositions) stable to biocorrosion; b) materials with antimycotic properties for prophylaxis and treatment of mycosis; c) biologically active polymer materials for protection of archaeological and museum exhibits; d) for human protection during their contact with microorganisms. Preliminary investigations have shown that the synthesized compounds have also a real perspective to be utilized as accessible antioxidants towards the cancer.

Chapter 21 - Quaternary arsonium triiodides $[(\text{Ph}_3\text{AsCH}_2\text{I})\text{I}_3]$ and $[(i\text{-Bu})_3\text{AsCH}_2\text{I}]\text{I}_3$ have been synthesized and studied.

The x-ray structures of $[(\text{Ph}_3\text{AsCH}_2\text{I})\text{I}_3]$ and $[(i\text{-Bu})_3\text{AsCH}_2\text{I}]\text{I}_3$ have been determined. Crystals belong to the monoclinic (comp.1) system, space group P 2₁/n (No. 14) with $a = 10.97$ (1)Å, $b = 13.152$ (1)Å, $c = 16.882$ (1)Å, $\beta = 93.01$ (1)° and to the triclinic system (comp.2), space group P-1 (No. 2) with $a = 8.413$ (1)Å, $b = 9.109$ (1)Å, $c = 15.876$ (1)Å, $\alpha = 76.24$ (1)°, $\beta = 75.60$ (1)°, $\gamma = 75.26$ (1)°. The structures were refined to an R value of 0.063 from 4082 (comp. 1) and 0.091 from 4475 (comp.2) observed reflections. The As atom is coordinated tetrahedrally to the substituents and the anion has a linear structure. The synthesis of $[\text{R}_2(\text{R}')\text{AsCH}_2\text{I}]\text{I}_3$ (where $\text{R} = \text{R}'$ or $\text{R} \neq \text{R}'$) are described.

The possibility of the perspective application of synthesized compounds has been shown.

Chapter 22 - Pollution of the environment is caused by human industrial and agricultural activity. It poses a harmful factor to the genetic apparatus of organisms to which are connected hereditary diseases, premature aging, cardiovascular problems, etc.

Numerous experimental investigations have shown that many chemical factors are characterized by a mutagenic influence. Following these discoveries, the author's laboratory elaborated many prophylactic measures, directed at prevention of genetic damage from the influence of harmful mutagenic agents on the organism.

In the laboratory, the antimutagenic and anticytotoxic effect of bioenergoactivators (biorag, ragozan, ematon and ragil) on mice have been studied during mutation and cytotoxicity induced with the fertilizer ammonium nitrate and the pesticides (phosphamide, trichlorfon and celtan). The cytologic and genetic methods of investigation have been applied in the study. Experiments showed that tested bioenergoactivators exerted highly effective automutagenic and anticytotoxic action.

Environmental pollution, which basically is caused by agricultural and industrial activity of human beings, comes back to them as factors harmful to organisms and their genetic apparatus, resulting in not only hereditary illness, malignant tumors and premature senescence, but also illnesses such as cardiovascular and digestive system disease, neural, allergy, and others.

In connection with an annual increase of chemical pollutants, science stands in the front of genetic danger. Among those problems whose solutions are first and foremost, the protection of organisms and their progenies from chemical mutagens existing in the environment is the most actual one. Wide application of chemical preparations in medicine, agriculture, industry and everyday life, as well as the existence of a great amount of chemical pollutants in the soil, water and atmosphere, allow us to talk about a sharp alteration of the ecologic situation [1].

Chapter 23 - This article is dedicated to the 100 years anniversary of birth of Georgian prominent scientist, Professor Akaki Gakhokidze. He is one of outstanding representatives of Georgian chemist's school. High theoretical preparation, mastery of experiment conduction, unusual scientific flair and intuition allow him to leave the great and light footprint for the posterity on the way of scientific research and pedagogical activity. Fundamental investigation of A.Gakhokidze won the international recognition. His works were published and broadly considered in the special literature, monographs and manuals of chemistry. There are created the specific paragraphs as "Synthesis of Gakhokidze", "method of Danilow-Gakhokidze" etc.

**PART 1. CHEMISTRY, USE AND MOLECULAR
MODELLING OF BIOLOGICALLY ACTIVE
COMPOUNDS**

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