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Otto Vogl

University of Massachusetts - Amherst, vogl@polysci.umass.edu

Josef Schurz

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Polymer Science in Austria: Universities in Vienna

Josef Schurz^a and Otto Vogl^b

^aInstitute of Physical Chemistry, University of Graz, A-8010 Graz, Austria

^bPolytechnic University, Six MetroTech Center, Brooklyn, NY 11201, USA

in cooperation with Oskar F. Olaj, Gerhard Greber and Werner Praznik



Josef Schurz



Otto Vogl

Vienna has eight universities: the University of Vienna, the Technical University of Vienna, the University of Economy of Vienna, the University of Veterinary Medicine, the University of Agriculture (Universitaet fuer Bodenkultur), the Academy of Arts, the University of Applied Arts and the University of Music and Applied Art.



Vienna with Minorite Church and City Hall.

UNIVERSITY OF VIENNA

The University of Vienna, the "Alma Mater Rudolphina," has about 60,000 students and 8 faculties: Catholic Theological Faculty, Protestant Theological Faculty, Faculty of Law, Faculty of Social and Economic Science, Faculty of Medicine, Faculty of Basic and Integrative Science, Faculty of Philosophy, Faculty of Formal and Natural Sciences.

From over 2000 universities in the world, the University of Vienna is considered the 17th oldest university. It was founded in 1365 by Duke Rudolf IV. In the 15th century it had about 1,000 students and was one of the leading universities in Europe. During the centuries it had its ups and downs, reflecting the general struggle between church and state. In the last two centuries, it has developed strong affiliations in philosophy and medicine.



University of Vienna, main building.

The first building of the Chemical Institute was built in 1872; subsequently, the new main building of the University, situated on the Ringstrasse of Vienna, was constructed in 1884. In 1915, a new complex of Institutes, including chemistry, physics and mathematics, was built on the Währingerstrasse. It remains the home of chemistry, physics and mathematics at the University.

Since 1365, almost 1 million students have attended the University and some 180,000 have received academic degrees. More than 5,000 scholars have been professors at the University.

The Institute of Physical Chemistry (Oskar F. Olaj)



Chemical Institute, University of Vienna.

Polymer research has had a long tradition at the University of Vienna. It began in 1932, when Herman Mark was appointed Professor at the University of Vienna and became the head of the so-called I. Chemical University Laboratory. During Mark's teaching period at the University, fundamental work was carried out on free radical polymerization, rubber elasticity and on a number of other projects which became part of the reputation that we associate with polymer science at the University of Vienna.

Mark surrounded himself with a number of young, talented and highly competent scientists. Among them were Franz Patat, who became Professor at the University of Innsbruck and later at the Technical Universities of Hannover and Munich. Patat played a significant role in the development of coordination insertion polymerization. Otto Kratky, another of Mark's assistants, became Professor at the University of Prague and at the University of Graz; he was the inventor of X-ray small-angle scattering. Other scientists of the Mark school were Eirich, Guth, Simha and Broda.

Johann Wolfgang Breitenbach remained at the University of Vienna and eventually was appointed to the chair of physical chemistry that Herman Mark formerly occupied. He expanded teaching and research in polymer chemistry and held the chair for more than three decades. Among the numerous topics that he and his group investigated were the kinetics of free-radical polymerization, especially chain transfer reactions, spontaneous polymerization, inhibition of polymerization, popcorn (inhomogeneous)



Strudelhofstiege.

polymerization and copolymerization, electroinitiated polymerization, phase-separation studies of polymer solutions, polymer fractionation, light scattering and osmometry of polymer solutions. Almost 300 scientific communications are witness to this prolific epoch at the Institute of Physical Chemistry.

In the sixties, the I. Chemical University Laboratory of the University of Vienna (at that time already named the I. Chemical Institute) was divided into an Institute of Inorganic Chemistry and the Institute of Physical Chemistry. Polymer Chemistry, because of the physicochemical character of research work done under Breitenbach, remained at the Institute of Physical Chemistry.

In 1977, Oskar Friedrich Olaj was appointed Professor of Chemical Physics and succeeded Breitenbach. He was also elected head of the institute, a position which he held until 1984. New professorships were created at the Institute of Physical Chemistry and Olaj is now one of the four (full) Professors of the Institute.

Olaj had originally started as a kineticist with a thesis on multifunctional chain-transfer agents (carbon tetrabromide and hydrogen sulfide) for radical polymerization under Breitenbach. For a number of years, he worked on electroinitiated free radical and ionic polymerization and also on several topics of free radical polymerization. He investigated the spontaneous, thermal initiation of styrene and σ -chlorostyrene polymerization.

In 1971, Olaj extended his interest to statistical thermodynamics of polymer systems, studied by computer (Monte Carlo) simulation. These two general topics, computer simulation of polymer systems on the one hand and polymerization kinetics on the other hand still are the main fields of polymer research at the institute. In the following sections, some actual topics of the work that is being done under the direction of O.F. Olaj will be described.

Computer simulation of polymer systems (Gerhard Zifferer). This work deals with single-chain properties—the evaluation of macroscopic (global) and microscopic (local conformations) properties as a function of solvent quality, chain-pair properties

Centers of Polymer Research

such as pair-distribution function and chain dimensions as a function of chain-separation and concentration, again also under the influence of the solvent quality.

Quite recently, these studies have been extended to the investigation of the behavior of chains close to a hard wall. Simulation of dense polymer systems is also being investigated. Remarkable progress was made when new quasi-kinetic relaxation mechanisms were introduced which were no longer bound to the presence of vacancies. The most outstanding result gained by this method was the confirmation of Flory's theorem (chain dimensions in bulk are equal to those in the unperturbed state). This procedure has now been successfully extended to phase-separation phenomena between two polymers of limited compatibility, to the simulation of the interface between two such polymers and to the evaluation of the concentration profile and the chain-size distribution across the interface. Another project involves the properties of a single polymer chain (type A) embedded into a matrix consisting of a thermodynamically different polymer (type B) and of a solvent.

Kinetic investigations of free radical polymerizations with the objective of determining individual rate constants (Irene Schnoell-Bitai). Rate measurements in stationary state polymerization or measurements of the degrees of polymerization, when the polymer is formed, invariably gave the rate constant of propagation k_p and the rate constant of bimolecular termination k_t in the combination k_p/k_t with no chance of separating these two contributing factors. Resolution into the components of the ratio usually was made by resorting to pseudostationary rate measurements (i.e., under discontinuous, however periodic initiation, e.g. the rotating sector method) which yielded k_p and k_t by obtaining the ratio k_p/k_t .

Not only was this method successfully applied to homo-

polymerization (e.g. of styrene, styrene-derivatives, methacrylates, acrylates, etc.) but it could also be adopted for the kinetic analysis of copolymerization reactions (styrene-methylmethacrylate, styrene-methyl acrylate); k_p and k_t could be obtained as a function of the monomer feed compositions. The surprising conclusion was that the well accepted Mayo's terminal model was found not to apply here for k_p . Efforts are now underway to further develop new methods of pseudostationary polymerization, to apply them to monomers which have hitherto escaped the determination of k_p and k_t , and to apply them to terpolymerization.

Theory and simulation of polymerization processes (Gerhard Zifferer). A third important research area which is being investigated with Associate Professor Zifferer involves the theoretical description of various polymerization processes and the evaluation of the chain-length distributions of the resulting polymers. This field is in part connected with the problems raised in polymerizations in the pseudostationary state where closed solutions of the differential equation systems could be worked out only in the simplest cases (pulsed laser initiation and rotating sector initiation).

In order to overcome these shortcomings, a versatile method has been developed which directly simulates the development of the pseudostationary state and finally yields the numerical solution after a few steps of the time-profile of the radical concentration. This profile then serves as the basis of calculating the average rate of polymerization and the chain-length distributions. The surprising result is that fully arbitrary periodic initiation profiles could be used for the establishment of pseudostationary conditions, suitable for the evaluation of k_p and k_t along the lines described before.

Excitation energy transport and energy transfer. Another independent branch of research under the leadership of Associate Professor Harald F. Kauffmann is concerned with the elucidation of the nature of optical excitations in aromatic polymers by photophysical methods, in particular by time-resolved fluorescence.



Vienna, St. Stephens Cathedral.



Schonbrunn Castle, Vienna.

This technique has not only developed into a powerful tool of probing transient excited-state events, but it is also very helpful in the phenomenological detection of relaxation processes, typically on sub-nanosecond and nanosecond time scales. In addition, transient fluorescent studies have provided evidence for the presence of excitation energy transport (EET) and direct energy transfer (ET). Thus, at least in principle, information on both static and dynamic polymer properties is accessible.

The main focus of this work was concerned with aromatic polyamides. Unfortunately, even homopolymers represented such a highly complicated system that it was impossible to provide a quantitative translation of fluorescence data into specific parameters of polymer structure on a microscopic molecular level, mainly because of the deficiencies of the theoretical basis available.

Current efforts in the group, therefore, concentrate on computer simulation experiments which are helpful in bridging the gap between the microscopic treatment of the polymer-inherent EET-many body problem on the one side, and the ensemble averaged fluorescence-observables on the other. Both the synthesis of polymer fluorescence data via the Monte Carlo-master equation routines and the analysis of such data (reconstruction of distributions) with the help of unbiased regularization provided an important technique of probing different conformational topologies and photophysical situations in these polymers. Accordingly, they make a considerable impact in the interpretation of polymer fluorescence in a real data experiment.

Wood research. Austria is a very important wood producing country. Consequently, research on cellulose and on the other important wood component, lignin, has a long tradition in Austria. In the middle 1930s, Professor Anton von Wacek started his research on lignin and other wood components. He identified pyrogallol derivatives in hard wood and also started to explore the possible use of lignin products as components for synthetic resins. His coworker, Karl Kratzl, continued and greatly expanded the work on lignin and studied the acid, alkaline and oxidative degradation of lignin. Other substances present in wood were also investigated such as talloil, natural resins and terpenes. At the present, no further work is being done in Vienna on this most important renewable resource—wood.

Technical University of Vienna

The Technical University received its present name in 1975. Over a period of almost 300 years it underwent many evolutionary changes. The first institution, which became one of the predecessors of the Technical University of Vienna, the "Engineering Academy," was founded in 1717. In actuality, Prince Eugen, who delivered Western Europe from the threat of the Ottoman aggression, proposed the creation of two engineering schools, one in Vienna and one in Prague; the education in these institutions was mainly aimed at military training.

The direct root of the Technical University of Vienna can be found in the "Polytechnical Institute" which was the successor of the Engineering Academy. The Polytechnic Institute was opened in 1815 and was one of the oldest of its kind, the others being in Paris and Prague.

The Polytechnical Institute had a technical and a commercial department as well as a preparatory high school. In 1865, an extensive reorganization of the Institute was undertaken, the commercial department was eliminated and the Institute was



Vienna Opera.

renamed Technical College for Advanced Technology. In 1901, this Technical College for Advanced Technology of Vienna obtained the right to graduate their candidates with a degree of doctor of engineering and the first doctor of technical science graduated in 1902.

Teaching and research in macromolecular science is carried out in the department for Chemical Technology of Organic Compounds.

The Institute of Chemical Technology of Organic Compounds (Gerhard Greber)



G. Greber



H. Gruber

When the Polytechnical Institute of Vienna was created, two teaching chairs were established for technical chemistry, one for "Common Technical Chemistry" and the other one for "Special Technical Chemistry." In 1866, the latter was changed into what we know today as the "Institute for Chemical Technology of Organic Substances."

Polymer research and education at the Technical University started in 1976, when Gerhard Greber was appointed Director of the Institute of Chemical Technology of Organic Materials. The research activities were oriented predominantly toward preparative polymer chemistry. Since 1980, the work has been carried out in close cooperation with Associate Professor Hans Gruber. The research activities cover a broad range of topics on both synthetic and natural

Centers of Polymer Research

polymers.

High-temperature Resistant Polymers research is carried out on new aromatic polyamides, polyamides and polyimides with high temperature resistance and good processibility. Block copolymers are also being prepared. By varying the chemical composition of the blocks and block length a variety of new products with "tailor made" properties, e.g. different glass transition temperatures and thermostabilities up to 450 °C, could be obtained which were also soluble and fusible. Block copolymers with good processibility and with thermostabilities of more than 500 °C could be obtained by the utilization of aromatic diamines with sulfide and sulfone groups as structural elements for the polymer blocks.

Crosslinked polymers. New crosslinked hydrophilic gels on the



Belvedere Castle, Vienna.

basis of glucose- and sucrose methacrylates have been synthesized. The degree of crosslinking was determined by the ratio of mono- to polyfunctional esters in the monomer mixture. The gels were found useful as polymeric carriers. Reagents or reactive groups could be fixed by esterification or etherification of the free HO-groups of the carrier. Specific examples are: mixtures of mono- and polyfunctional methacrylic esters of sucrose which were prepared by transesterification of sucrose with methyl methacrylate. Radical polymerization of the monomer mixtures yielded crosslinked hydrophilic gels swellable in water or in polar organic solvents. They have potential use as polymers with: chelating groups; groups with catalytic activities; Ruthenium-bipyridyl complexes suitable for photochemical reactions; Girard-P and Girard-T reagents to detect and/or remove aldehydes and ketones.

Polymeric catalysts as enzyme models. Polymer models mimicking the active site of the enzyme α -Chymotrypsin with spacer groups have been synthesized. The activity of the enzyme models was found to be 25 times higher than other known models and the activity could be maintained for months without substantial decrease. The native enzyme α -Chymotrypsin still exhibits an activity eight times higher than the new polymer bound enzyme model.

Polymers for medical uses. Work in the department deals with the improvement of the tissue compatibility of silicone rubbers.

Polymer additives. This type of research involves the study of novel additives with low volatility, good compatibility with organic materials and stability against migration. The additives that were

studied were: telechelic antioxidants, light stabilizers and optical brighteners for polyolefins based on polyethylene oligomers with covalently bound additive end groups; thermal stabilizers for PVC based on organotin compounds with highly chlorinated aliphatic groups; modified silicas have been synthesized by the reaction of silica with methylchlorosilane and subsequent hydrosilylation reactions.

Photoinitiators. New photoinitiators with reactive groups (e.g., hydroxy-, amino- epoxy-groups) or polymerizable groups on the basis of hydroxyalkylbenzophenones have been synthesized. Polymeric photoinitiators with "tailor made" properties could be obtained by radical polymerization of the polymerizable initiators or by copolymerization with vinyl monomers.

Natural polymers. A new process for the preparation of regenerated cellulosic fibers has been developed by silylation of cellulose with trimethylchlorosilane and subsequent hydrolytic cleavage of the trimethylsilyl-cellulose.

Natural raw materials. Research projects on natural renewable materials deal mainly with the preparation of new sucrose derivatives. Without changing the basic structure of sucrose, a number of new sucrose ethers and esters containing functional groups have been synthesized. These derivatives are not single, well defined compounds, but rather mixtures of isomers with various degrees of substitution. Examples are: sucrose methacrylates; sucrose derivatives with primary amide groups of N-methylolated amide groups as condensation components for urea- melamine- and phenolic resins. The properties of these sucrose- -amidocetyl ethers and of the amino- or phenolic resins prepared therefrom could be varied by suitable inert substituents to be either hydrophilic or hydrophobic; polyfunctional sucrose derivatives with aromatic azide groups as photoactive curing agents for the preparation of photo-resists with good adhesion to metallic surfaces; sucrose derivatives with primary amino groups and their fatty acid amides with surface active properties—these compounds act as detergents with higher hydrolytic stability than the known sucrose fatty acid esters; water soluble sucrose derivatives with biologically active groups, e.g. esters of sucrose with acetylsalicylic acid.

Telechelics. Telechelics with hydroxy, amino or carboxyl end groups have been synthesized by anionic polymerization of styrene or butadiene and by termination with suitable functional reagents. Radical polymerization with functional initiators are also being studied.

Silanes and polysiloxanes. Silanes with photoactive dimethylmaleimide or azide groups bonded by hydrolytically stable Si-C bonds are being modified by hydrosilylation reactions of hydrogen silanes. These polymers are then converted to functional polysiloxanes for their potential use as photoresist materials.

Photocrosslinkable alkyd resins. New photoresists with dimethylmaleimide or azide groups have been prepared by condensation polymerization of polyfunctional alcohols with suitable functional phthalic anhydride derivatives.

Graft copolymers. Fiber forming graft copolymers based on cellulose or cellulose acetate were synthesized by chain transfer polymerization using thiol group containing cellulose derivatives as comonomers for styrene, methyl methacrylate, acrylonitrile and hydroxyethyl methacrylate copolymerizations.

Teaching at the Institute. About 50 M.S. and 40 Ph.D. students have graduated in polymer chemistry from the Institute since 1976. The following courses are available at the Institute: "Introduction to Chemical Technology of Organic Materials," "Chemical Technology



Peterskirche, Wien.

of Organic Materials I; "Chemical Technology of Organic Materials for Chemical Engineers" and a laboratory course "Chemical Technology of Organic Materials;" a seminar is being presented by experts from the chemical industry.

UNIVERSITÄT FUER BODENKULTUR

The Universität fuer Bodenkultur, the "Alma Mater Viridis," was founded in 1872. It now has 7300 students, 260 professors and assistants. The scientific research programs are concerned with agricultural, forest-technological and biotechnological subjects. All the polymer related research is being carried out in the Institute of Chemistry, which is headed by Professor Leopold I. Maerz.

Institute for Chemistry (Werner Praznik)

The work in the Institute of Chemistry is concerned with the optimal utilization of renewable biological raw materials combined with the maximum preservation of ecological circles. This approach is considered one of the most important tasks of bioscience.

Extensive basic research and increased development is being carried out for the application in biotechnology. Biopolymer research is being done by three working groups, the most important research group in carbohydrate research is headed by Werner Praznik.

Two other groups are engaged in research of glycoconjugate. The group of L.I. Maerz is concerned with the characterization of complex carbohydrates and the investigation of their interactions with biological systems—bacteria, plants, animals and man. Glycoproteins from insects are being investigated with major emphasis on the structure and biosynthesis of their carbohydrate side chains. It is expected that immunogenicity or antigenicity could be created, when aglycoprotein is administered to mammalian



W. Praznik

organisms.

Organic synthesis of complex C- and polysaccharides is the main objective of the work being done in the group of P. Kosma. Their interest is focused on a specific lipopolysaccharide (LPS) of a gram-negative bacteria, also known as endotoxin. The various segments of this complex compound are associated with distinct biological effects. The objective here is to synthesize an artificial, toxicologically harmless immunogen. This work uses extensively NMR spectroscopy, studies of molecular conformations and molecular modeling.

The group of W. Praznik is working on polysaccharides from renewable plant raw materials, especially those which are of interest for biotechnological utilization.

Starting with the general interest in potato starch, further research into the domain of starch led to the separation of potato starch into different molecular weight fractions. Separation of amylose and amylopectin was possible by chemical complexing techniques and by means of size exclusion chromatography.

High molecular linear glucans have been produced by the catalytic action of phosphorylase utilizing malto-oligomers and glucose-1-phosphate (this work is being done in cooperation with B. Pfannmueller, Freiburg/Germany).

The isolation and characterization of fructans from various plants is also a significant part of the research efforts of this group; of special interest are the components of Jerusalem artichoke and chicory. These plants contain a high percentage of water soluble linear polysaccharides, fructans known as inulins, which could be utilized for the production of high class fructose molasses. One of these fructans, sinistrin, has a branched structure and is a water soluble fructan; it is now used in medical applications for a renal clearance test.

A close cooperation with the Institute for Physical Chemistry of the University Graz (J. Schurz, O. Glatter, A. Huber) has existed for several years in the field of biopolymer characterization. A clear correlation between molecular polymer characteristics and macroscopic material properties seems to exist. Such parameters provide basic information for successful biotechnological utilization (cooperation with K.D. Klube and E. Berghofer of the Institute for Nutrition Technology, Austria; with R.H.F. Beck of Cerestar, Belgium; and with R. Wutka of Laevosan, Austria). Cooperation in the agronomy of polysaccharide containing plants exists now with teams working on plant breeding (P. Ruckebauer) and on plant physiology (G. Soja of the Research Center of Seibersdorf, Austria).

The study of the structure of wood is an additional field of the

Center of Polymer Research

research of this group. Investigations are focused on the carbohydrate matrix of wood as a function of storage and/or drying. Work is also being done on the behavior of the individual types of wood under the normal conditions of drying. Glycoconjugates (glycolipids, glycoproteins, proteoglycans) occur in all pro and eucaryotic organisms, in soluble form as well as associated to cell surfaces. The physicochemical properties of the carbohydrate moieties lead to characteristic and often essential interactions in vivo and in vitro. Many of them are being investigated as recognition phenomena, among them infectious invasion of tissues or such fundamental processes like contact inhibition or metastasis.

AUSTRIAN ACADEMY OF SCIENCES

The Austrian Academy of Sciences is an organization consisting of almost five hundred scholars as members. It has two divisions, the mathematical-scientific division and the philosophical-historical division. The Academy has 115 full members and is guided by a directorate of four. The president of the Austrian Academy of Sciences is Werner Welzig; Otto Hittmair is Vice President, Karl Schloegl is General Secretary and Walter Selb is the Secretary.

As early as 1713, the famous mathematician and founder of the

Prussian Academy, Leibnitz, suggested the foundation of an academy in Vienna, but it was not until 1847 that it was formally created. In 1857, the Academy moved into the main building of the old university. Over the years, the objective of the Academy was redefined and it ultimately became the leading research institution of Austria. Many new institutes were created and the cooperation with international organizations was initiated and/or organized.

In the mathematic-scientific division, many of the research activities are concentrated in institutes. The main emphasis is in physical, biological, medicinal and ecological sciences. One institute, the Institute of Solid States, is investigating the properties of technical materials using physical methods, particularly plastic deformation, fracture behavior, recrystallization and surface behavior.

Another institute is involved in the study of biologically active proteins, gene regulations, gene transfer and the molecular basis of muscle and cell movement. The focus of new activities is the interdisciplinary cooperation of technical, natural and social sciences with a special emphasis on the development of new technologies and their effect on society.



Austrian Academy of Sciences.