Proceedings of the Fifth International Congress on Catalysis, Miami Beach, Fla., 20-26 August, 1972

CATALYSIS

JOE W. HIGHTOWER

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edited by

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PREFACE

The Fifth International Congress on Catalysis was held in the USA at Palm Beach, Florida, August 21-25, 1972. Co-sponsored by the International Union of Pure and Applied Chemistry, the Congress was organized and hosted by The Catalysis Society of North America. Professor Paul H. Emmett was Honorary Chairman, while Dr. Vladimir Haensel served as Executive Chairman. Chairmen for the Sub-Committies included J. E. McEvoy (Advertising and Publicity), F. G. Ciapetta and J. G. Larson (Financial and Budget), Ms. G. A. Mills (Ladies' Hospitality), W. H. Flank and Ms. R. A. Svacha (Meeting Arrangements and Social Functions), A. E. Hirschler and A. Farkas (Paper Review), R. L. Burwell, Jr. and W. K. Hall (Paper Scheduling), J. W. Hightower (Publications), W. K. Leaman (Registration), J. G. Larson (Treasurer), and H. Heindmann (Grants).

Although originally scheduled for New York City, the organizers moved the recing to Miami, Florida, to avoid the congestion and high prices in NYC during summer months. When the President's political party switched its national convention to Miami at the same time, the meeting site was again changed to avoid any possible conflict; all agree that the change to Palm Beach, Florida, was a very wise decision. The elegant facilities of the Breakers Hotel provided a dignified and relaxed atmosphere conducive to maximum enjoyment of the Congress. Many of the approximately 500 delegates from 32 countries took advantage of the lovely Florida weather by sunning themselves or swiming in the calm Atlantic Ocean. Group social events included a get-acquainted cocktail party and a congress banquet. These were supplemented by "hospitality suites" kindly provided by the several catalyst manufacturers.

The theme of the Congress was "The Science of Catalysis." In addition to 107 papers selected from nearly twice that many which were submitted, six invited lectures by outstanding internationally recognized leaders in catalysis rounded out the program. Preprints of all papers were sent to the delegates six weeks before the meeting. Except for the invited lectures, the papers were presented in two parallel sessions with thirty minutes allocated for each paper—15 minutes for presentation and 15 minutes for discussion. Each commentator was asked to submit his oral question or comment in writing immediately after the session. These were then collected and sent to the authors for their written response. Among the papers which generated considerable excitement among the delegates were those on particle size effects, partial oxidation, supported homogeneous catalysts, and mechanism of reactions over pure or mixed oxides. The complete manuscripts, comments, and responses are included in the two volumes of the Proceedings.

The Sixth International Congress will be held in London, England, during the summer of 1976; Professor C. Kemball will be the organizing chairman. Officers of the International Congress include Professor G. K. Boreskov - President, Professor C. Kemball - President Elect, Professor R. L. Burwell, Jr. - Vice President, Dr. D. A. Dowden - Secretary, and Professor J. Haber - Treasurer.

The only sadness associated with the meeting surrounds the untimely death of three men who have been leaders in the International Congress for many years. Pictures and biographical information about each follow.

About 150 people actively participated in organization of this meeting. We are all indebted to them for their splendid efforts. The Editor would particularly like to thank A. Farkas for writing the Necrology section and helping edit several of the papers. He also thanks others on the Publications Committee (R. L. Burwell, Jr., T. R. Hughes, J. V. Kennedy, and J. T. Richardson) for their editorial help. Finally, our deep appreciation is due Mrs. Ruby Rost who did all the typing of the discussion and several manuscripts in their camera-ready form.

The personal contribution of \$10,000 by W. Clement Stone, a noted Chicago Philanthropist, made it possible for a substantial number of students and young instructors to participate in the Congress. Other financial supporters for which we are grateful are included in the lists of sponsors and

contributors which follow.

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The North-Holland Publishing Company (Amsterdam) has agreed to publish the Proceedings. The Organizing Committee expresses their appreciation to the managing editor, Drs W. H. Wimmers, for his preparation and shipment of both the Preprints and the Proceedings.

Houston, Texas January 10, 1973

Joe W. Hightower Editor of Proceedings Jan Hendrick de Boer

1899-1971



Jan Hendrick de Boer was born in Ruinen, Holland, in 1899. He received his Ph.D. in Groningen and joined Philips Gloeilampenfabriek in Eindhoven. His ingenuity in developing technological applications for scientific findings was soon recognized, and he was rewarded by promotion to laboratory head. The war forced Dr. de Boer to move to London in 1940 as director of the Dutch Laboratory of the Ministry of Supply - a position he held until 1945. From 1946 to 1950 he was head of the Unilever Laboratory in England, and then he became associated with Staasminen in Limburg. In addition to these industrial jobs, Dr. de Boer also taught at the Delft Technological University. In 1962 he was appointed president of the Scientific Council for Nuclear Energy. In the period 1964-71 Dr. de Boer was president of the International Congress on Catalysis. Dr. de Boer was one of those rare individuals who could perform brilliantly with equal virtuosity as a scientist, technologist, teacher or administrator, in both physics and chemistry, in any one of several languages, and whose technical knowledge was excelled only by his personal charm. In his research work he covered a dazzling variety of topics, all with superior competence and creativity. De Boer is the author of innumerable papers and patents on the separation of hafnium and zirconium, color centers in solids, photosensitive layers, fluorescence, phosphorescence, photoelectric tubes, rectifiers, electron emission, photographic materials, X-ray screens, sound recording and, of course, adsorption and catalysis. Dr. de Boer received many awards and honorary degrees and was elected to several academies. When he retired in 1969, his former students and coworkers dedicated a book to him entitled "Physical and Chemical Aspects of Adsorbents and Catalysts" - a truly remarkable record of the accomplishments of de Boer and of his school. But de Boer was not to enjoy his retirement for long. On April 26, 1971, he died, and the scientific community of the whole world mourns his passing.

Frank George Ciapetta

1915-1972



Frank George Ciapetta was born in Philadelphia in 1915. After receiving the B.A. and M.A. degrees from Temple University, he joined the Atlantic Refining Company in 1939. In 1946 as a du Pont fellow Frank returned to academic work at the University of Pennsylvania where he was awarded a Ph.D. degree in 1947. Then followed a most successful eight-year research campaign on the applications of catalysis to hydrocarbon chemistry, half of which was spent at Atlantic and half at Socony as a research associate. This work covered virtually all typical refinery reactions, including alyklation, isomerization, cracking and reforming, and culminated in Frank's receiving the coveted Precision Scientific Company Award of the American Chemical Society in Petroleum Chemistry in 1955. The same year Frank joined W. R. Grace & Co. as research director of the Davison Chemical Division where he was promoted to vice president of research in 1968. Frank's enthusiasm for, and success in, matters catalytic continued at Davison and led to some remarkable developments in cracking, auto exhaust and polymerization cata-Frank was the author of about seventy papers and patents and participated actively in many scientific and technical organizations. He was chairman of the Petroleum Research Fund Advisory Board and president of the International Congress on Catalysis when he suddenly died on February 7, 1972. Everybody who has known Frank will remember him for his technical accomplishments and his leadership qualities; in his passing we mourn the loss of a true friend.

Alfred Ernest Hirschler

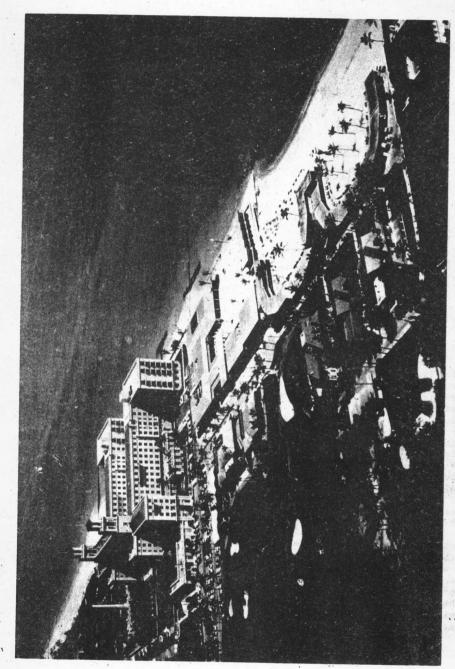
1911-1972



Alfred Ernest Hirschler was born in 1911 in Bluffton, Ohio. After graduating from his hometown college in 1933, he went to Ohio State for his Ph.D. which he recieved in 1937 .. The same year he joined the Sun Oil Company as a research chemist. In 1940 he was promoted to group leader and became a research associate in 1960. Al's research work concerned solution thermodynamics, oxidation of hydrocarbons, fuel combustion, physical properties of hydrocarbons, separation and purification of hydrocarbons by adsorption, purity determination by freezing point measurements and cracking catalysts. Al devoted a number of years to the study of the acidity of these catalysts with particular emphasis on the correlation between acidity and catalytic activity and on the source and nature of acidity. The work on catalyst acidity became well-known and Al was recognized as an authority. Early in 1972 Al was selected to receive the Philadelphia Catalysis Club Award "for outstanding contributions to the advancement of catalysis." The award was to have been presented in May, but Al died suddenly on February 23, 1972.

Al was an active member of the American Chemical Society, the Philadelphia Catalysis Club and other technical organizations. The chairmanship of the paper review committee of the Fifth International Congress on Catalysis was Al's last professional assignment. As he did with all other tasks, Al accomplished this one too with methodical dispatch and exemplary efficiency. Al was a hard-working, reliable researcher with a sincere and forthright character. He was mild-mannered and modest, but stood firmly for his convictions and had no difficulty in holding his own in a scientific discus-

Al is sadly missed by his friends, coworkers and colleagues.



The Breakers Hotel - Palm Beach, Florida

OPENING REMARKS

<u>DR. V. HAENSEL</u>, Chairman of Organizing Committee, Fifth International Congress on Catalysis

Delegates to the Vth International Congress on Catalysis, it is a distinct pleasure to greet you on behalf of the Organizing Committee and the many workers who have given so much of their time and talent to what we hope will be a most successful conclave on catalysis.

During the last 12 months we have suffered substantial grief along the way. We have lost DeBoer, Ciapetta and Hirschler. At this time I would like the delegates to observe a moment of silence in memory of these untiring workers in catalysis and contributors to the success of catalytic congresses.

You are aware of the difficulties in finding a new home for the Vth Congress. As far as I know, this is the first Catalysis Congress which had to move on a moment's notice some 60 miles or 100 kilometers from its original destination, which we so carefully prepared some three years ago. In our world, there is room for science and politics as well as scientists and politicians, however, we did not wish to expose the catalytic scientist to the rougher elements which nowadays inevitably clutter up the arena of a political convention.

So now we are here, and, if some things are not exactly to your liking, please forgive us since we have tried to do our best. If you like them, tell us, we like to have a little praise for our efforts. Similarly, since we have had to select some 110 papers from nearly twice that number originally submitted, we faced some very difficult decisions in accepting some and rejecting others. It was not a choice between poor and excellent, but most of the time it was a choice among very good papers, some of which fitted the theme of the Congress somewhat better than others.

We hope that most of you have had an opportunity to read the preprints and to select the ones in which you would like to participate as a discussor or as a listener. Having done some of this homework myself I have made certain observations that I would like to share with you.

The theme of the Congress is the Science of Catalysis, and there is no doubt that these papers represent by and large some thought provoking contributions. The experimental procedures have become more elegant and thus the interpretations more intriguing. At the same time, one cannot avoid the thought that some of the work digs merely deeper in the territorial imperative, which is quite natural for someone who is the top expert in his own field. And yet it appears to me that these brilliant workers could contribute so much in other perhaps more relevant areas.

How does one go about finding these more relevant areas? This is where the gulf between the industrial and academic sectors is the greatest. The shroud of secrecy overhanging industrial research does a great deal not only to obscure the fine contributions already made in industry, but quite frequently it obscures the existence of the problem itself. It is no wonder that a recent editorial in "Science" pointed out that many professors convey

the impression to their students that applied industrial research is a second rate occupation for second rate people.

This Congress, through its communicative powers, can do much to bridge this gulf. It is essential that we do this in the spirit of trying to extend both the understanding and the application of catalysis. You will note in your program that we acknowledge in particular the gift from Mr. W. Clement Stone, who sends his best wishes for the success of the Congress, underscoring this Congress as an opportunity to provide solutions to problems of concern to humanity.

What are the problems of concern to humanity that catalysis may help in solving? So let us carve out the territorial imperative where we believe catalysis can help. The area of energy is a very pertinent one. Here we are concerned with improved methods of conversion of raw materials into products that we need. These raw materials are oil, coal, shale and tar sands. The products we need are gasoline, jet fuels, heating oils and synthetic natural gas.

Catalysis is deeply involved in these conversion systems, and most of you will be amazed to learn how much basic catalytic research is required to help resolve the applied problems. A number of these can be done at the university level if the members of the academic fraternity are willing to work on them. At the same time industry must be willing to share these problems with the academic society.

The same concept applies to another problem, this one connected with the use of the products derived from our raw materials. These products, as well as the raw materials, are burned and in the course of the combustion process we get into all sorts of pollution problems. Here you will be amazed at the amount of basic research work that needs to be done in conjunction with the catalytic processing schemes that have been proposed for these pollution problems. Because of the urgency of the pollution problem much of the basic work remains undone and, here again, industry needs the cooperation of the academia to undertake some very basic aspects of the applied problems.

Thus, the gulf is wide but can be narrowed by cooperation and giving on both sides. Right now, if you don't mind a facetious remark on my part, I believe that the academic and industrial fraternities are separated by one atom only. This is the nitrogen atom. The academic people are decomposing N2O while the industrial people are convering NO. Now if we can only get rid of that one additional nitrogen atom in the academic fraternity we shall be on the same wave length.

DR. G. ALEX MILLS, President of the Catalysis Society

It is my privilege on behalf of the 1,571 members of the Catalysis Society of North America to express appreciation to all those who have contributed to make this Vth International Congress on Catalysis a success-speakers, discussion leaders, attendees and organizers.

It is to the group or collective activities of those involved in the science of catalysis that I address you briefly. First, in this regard we must applaud Dr. Vladimir Haensel, Chariman of the Organizing Committee and his hard working Committee.

It is the naive belief of some that science can flourish solely by the action of individual scientists. But, while scientific discoveries are indeed accomplished by individuals, it is also true that much more can be accomplished by uniting for the advancement of certain types of important activities, namely those which involve exchange of ideas.

The exchange of information and ideas is the essence required for creativity. New concepts are usually arrived at by discussion and argument, frequently controversial and always exciting. Just inspect the wide range of references in a technical article. Now add to that the benefits of discussion with the author. How I wish we could measure and document the value of our Catalysis Society activities so as to demonstrate their importance and to guide us in functioning even more effectively.

Here in North America we have organized for action in catalytic affairs by forming the Catalysis Society. We do not have a Catalysis Institute as some countries do. Since the Catalysis Society is a unique organization, I hope that a brief review and some comments on our activities will be of interest to those from abroad as well as to members of the Society.

Evidently, we are doing something right because we have become increasingly active for nearly a quarter of a century, having grown in membership since the inception of the Catalysis Club of Philadelphia in 1949 and the formation of the International Congress on Catalysis in 1956. There are now approximately 1,600 members, located in eight clubs or local societies, seven in widely distributed locations in the United States and one in Canada.

No doubt one of the secrets is that we are a loose organization--loose in the sense that we have not been confined by rules and regulations, but have been concerned with the fundamental objective of providing a fellowship dedicated to the advancement of catalysis. In other words, our main concern has been with getting things done.

Another and even greater reason for the success of the Catalysis Society is the enthusiasm of a large number of our members. It is interesting to note that many of the same persons who were members of that original small group who founded the Philadelphia Club in 1949--23 years ago--are still active and are here tonight. I am happy to count myself one of them.

It occurs to me that catalysis is a sort of disease, and, in fact, an incurable disease because here are individuals bitten by the catalysis bug a quarter of a century ago who have not yet recovered. And in fact, even worse, they have in the meantime infected many others who have now caught this incurable disease. May I introduce five of those sort of beneficial "Typhoid Marys" who are here today from the original group.

Dr. Alex Oblad, Dr. Heinz Heinemann, Dr. Roland Hansford, Dr. Adalbert Farkas, and Dr. Charles Plank.

Now let us look briefly at what the Catalysis Society does. The principal accomplishment has been to increase knowledge and interest in catalysis by providing a means for dissemination of knowledge and interchange of ideas. This has resulted, I believe, in significant additional progress in both the

science and application of catalysis. (In monetary terms, catalytic discoveries in the last two decades have resulted in creation of many billions of dollars of wealth as well as making possible technology and products which would not otherwise have been possible). Increased interest has been possible chiefly through the local meetings organized by the 8 individual societies which have provided addresses by experts, discussion and personal contacts. These meetings are usually held monthly in addition to annual one or two day symposia.

The Catalysis Society has also contributed to the advance in Catalysis by its sponsorship of the International Congresses which provide for discussion on a world wide basis--29 nations are represented here. The Society has also held two North American meetings--with the third scheduled for San Francisco in February 1974. At the meeting earlier 1971 in Houston, Texas, Dr. Hightower, Chairman, reported an attendance of 300 people--a remarkable 20 percent of total membership.

The Catalysis Society is also proud of its sponsorship of a society lectureship which has just been named the Ciapetta Lectureship in honor of the late Frank Ciapetta. This Lectureship carries a modest honorarium and a suitably inscribed plaque. We feel the distinguished lecturers have brought us distinction. These have included: Dr. Boer, Dr. Teichner, and Dr. Frank Stone.

Our current lecturer is Dr. George Schuit and we want to publicly acknowledge our debt to him. I am happy to report that Dr. Eischens has recently proposed Dr. Fripiat Lecturer for 1972-1973, and he has accepted this appointment.

The Society has rendered a service in publications as well. Dr. Hightower made available, for example, the papers in English of the Fourth Congress held in Moscow.

It gives me a certain amount of personal pleasure to tell you that we have received a measure of academic recognition, having recently participated as a "learned society" in the inauguration of two University Presidents.

A further activity which is popular with members is the Catalysis Newsletter, issued 2 or 3 times a year, alerting all members to meetings and events. The first and also current editors are present: Jim McEvoy and Maurice Mitchell.

Recently, two substantial awards were arranged by Dr. Heinemann and his committee. The Paul Emmett Award in Fundamental Catalysis, sponsored by the Davison Division of W. R. Grace Company and the Eugene J. Houdry Award in Applied Catalysis sponsored by the Houdry Process Co., Sun Oil Co., Oxy Catalyst Co. and members of the Houdry family. These awards are for \$2,000 and \$2,500 respectively. We believe that the publicity and financial rewards will contribute to the advancement of catalysis. I would like to recognize the winners of the first awards: Dr. Herman Bloch (Houdry) and Dr. Richard Kokes (Emmett). Nominations for the second award are due September 1, 1972.

It is now necessary to say what we have failed to do. We members of the Catalysis Society are also members of a larger society—the American community. But, in my view we are not adequately relevant and responsive to the

needs of this greater society. As a consequence of not being relevant and responsive, we in the United States have seen a loss of support for the science of catalysis by both government and industry. In my opinion there is something wrong with a meeting on catalysis in 1972 which does not have a strong program dealing with the catalysis for prevention of pollution. And there are other areas where catalysis could have profound social benefits. I know that some of my colleagues will object to my viewpoint. But I want to emphasize that I am not advocating a meeting with papers full of empirical industrial testing data. What I am advocating is that we begin to apply science of catalysis to problems such as pollution prevention, problems of concern to our society. I call on each of you to re-examine your selection of scientific research to include this important factor of usefulness to our society. I can only regard this as an opportunity to make catalysis of greater importance.

In closing, I should mention that all the officers and most of the members of the Board of Directors will this year complete their four year appointment. We look to a new team with new ideas and vigor to make the Catalysis Society more relevant, more responsive, and an even greater force in our highly industrialized society. Thank you.

PROFESSOR G. K. BORESKOV, President, International Congress on Catalysis
This is the fifth time that we have assembled an international meeting of scientists working in the area of catalysis. It is parcitularly fitting that this fifth anniversary Congress is being held in the United States which hosted the first Congress in 1956.

In connection with this I would like to remind you that we owe the establishment of the International Congress to the initiative and energy and energy of our American Colleagues. A great deal of effort was required to bring together the scientists active in the area of catalysis and to transform the Congress into the truly international organization that it is today. Much of this credit belongs to Professor deBoer whose success was due both to his high professional standing and to his warm, friendly personality.

There is now no question about the viability and productivity of this international cooperative effort devoted to the study of catalysis. Catalysis has provided the scientific basis for technological progress in a variety of areas and has materially benefited humanity. Its potentialities are by no means exhausted. Catalysis can be a powerful tool for solving the problem of how to protect the human race and nature in general from the harmful effects of technological progress.

I would like to wish success to all the participants in the Fifth International Congress on Catalysis, and I hope that this meeting will promote further international cooperation in this area, will speed up the theoretical understanding of this interesting phenomenon and will also lead to further practical utilization of catalysis for the benefit of all humanity.

PROFESSOR PAUL H. EMMETT, Honorary Chairman, Fifth International Congress on Catalysis

As honorary chairman of the program for the Vth International Catalysis

Congress it has fallen to my lot to say a few words about the future of catalysis. I do so with a full realization that I am not a prophet and that the pathways ahead are confused and not easy to follow. The most one can hope to do is to point out a few of the sign posts that have put in an appearance and call attention to the direction to which they are pointing. 1)

It takes little imagination to see that catalysis is entering an age of automation and computerized operation. Already surface area, pore sizes, pore distribution and in a few cases complete reaction rate measurements are all carried out automatically with little attention from the operator. This will become more and more the common thing rather than the exception.

There seems to be a certainty that the recent upsurge of interest in homogeneous catalysis and particularly in the catalytic activity of metallic complexes is destined to throw a great deal of light on heterogeneous catalysis. Indeed some of the papers that will be given at the present meeting show that progress is being made in actually fixing the inorganic complexes on to a surface in such a manner as to produce a heterogeneous catalyst having the same basic characteristics as the homogeneous catalyst but in addition much greater stability and flexibility. This work also points to a much better understanding of the detailed nature of the metallic complexes that are essential components of our body catalysts, enzymes.

Methods for utilizing controlled beams of x-rays, electrons, positive ions, neutral molecules and photons in surface studies are multiplying so fast that it is difficult to keep abreast of new developments. 2) In the past we have hoped that our catalytic surfaces were of a known degree of cleanliness but we had no way of being sure. Today with surfaces prepared at 10-10 mm pressure, analyzed for general conformation by low energy electron diffraction, and assured of purity by Auger spectroscopy, one, for the first time, has an opportunity to associate types of catalytic reactions with the inherent properties of clean surfaces. If promoters or alloys are being used it becomes increasingly possible to differentiate between the composition of the surface of the catalyst and its interior. In short a trip to some of the present laboratories that are focusing these new techniques on the activity of surfaces as catalysts impels one to expect a much more definitive appraisal of the effect of chemical composition and surface topography on catalytic behavior. It is safe to predict that progress, in this approach, will be fast and very revealing.

It augers well for the future when papers begin to appear in which theoretical calculations show relationships between the orbital characteristics of the reactant molecules and those of the catalyst surface. Two such examples come to mind. The first is a paper by Mango and Schackschneider³) in which they pointed out that the surface orbitals of transition metal oxides should make them excellent catalysts for the disproportionation reactions. A series of these reactions had been described by Banks and Bailey⁴) and later by Bradshaw, Howman, and Turner.⁵) A second example is to be found in the calculations by Ruch⁶) which show that the molecular orbitals of the nitrogen molecules are such as to make molecular chemisorption of nitrogen on iron surfaces reasonable. Furthermore, they showed that the 111 face of

the iron would be more capable of adsorbing nitrogen molecules than the 100 or 110 planes in agreement with the claims that 111 face of iron is many times more active for ammonia synthesis than the 100 or 110 planes.

Catalysts have often been referred to as the "heartbeats" of our industrial chemical processes. Probably the biggest application has been in the field of petroleum refining. With the advent of our present antipollution activities catalysis seems destined to gain prominence in at least two new areas. It will be essential in preparing high octane fuels to replace some of the additives that seem likely to be banned from gasoline products of the future. Secondly, catalysis appears to be an essential part of any program to eliminate carbon monoxide, hydrocarbons, and nitric oxides from the automobile exhaust gases. Expansion of its use in space exploration also seems likely. In a word catalysis of the future is likely to increase to the point of being even more vital to our existence than it has been in the past.

In a word, I am optimistic about the future of catalysis because of the many new research tools and approaches that are being introduced, because of the increasing interest in basic work both in homogeneous and heterogeneous catalysis and because of the ever increasing industrial demand for new, different, and better catalysts to help solve the problems that are confronting us. However, I am optimistic most of all because of the fine group of young scientists throughout the world who are tackling with enthusiasm and skill the many problems that we of an older generation have left unsolved.

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