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CURRENT PROBLEMS IN SCIENTIFIC-TECHNICAL CO-OPERATION IN
CATALYSIS BETWEEN A RESEARCH INSTITUTE AND INDUSTRY IN YUGOSLAVIA^{1/}

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INTRODUCTION

Chemical industry in Yugoslavia is characterized by a long tradition but with nonuniform development also, with periods of stagnancy and disproportion in the development of the particular branches.

Discontinuity in the development of the chemical industry reflected itself upon the development both of the fundamental and applied research in the industry itself and upon the collaboration with the scientific institutions engaged in the work for industry. The lack of a broader interest for the technological and economic aspects of the catalysis in industry and the neglect of the scientific research are particularly remarkable in this domain.

During the past decade this situation substantially changed. The complex investigations in the catalysis in a scientific institute linked tightly with several large enterprises, using catalytic processes, emphasized a whole series of problems of common interest. This collaboration is planned to be more intensive in the future as the role of the catalysis is growing up more intensively, particularly in the future development of the basic organic chemical industry.

Scientific-technical collaboration in the domain of catalysis in Yugoslavia will not be restricted to the existing contacts: institute - industry, but more and more will include the exchange of the achievements at the international level. On the basis of the past and future development of the chemical industry and already obtained results in the scientific-technical collaboration in the field of catalysis as well, may be concluded in which direction the international collaboration in catalysis is to be oriented and what are the problems to be solved by the united efforts.

III. THE DEVELOPMENT OF THE BASIC CHEMICAL INDUSTRY IN YUGOSLAVIA.

The first capacities of the basic chemical industry were installed on the territory of Yugoslavia in the second half of the nineteenth century, among which the Chemical and Electrochemical plants, the factories of sulphuric acid, hydraulic cement, oil soda etc. The period between 1900 and 1914 was characterised by the continuous development of sulphuric acid, calcium carbide, superphosphate and other chemicals production. All these plants in view of the capacities and applied technologies were not similar production in other countries.

In the period between two world wars the chemical industry of Yugoslavia, was developing under the strong influences of the foreign capital, and did not reach the expected level in spite of the previous tradition, very convenient raw materials and energetic possibilities, cheap labour and the requirements of domestic market. The growing of a great number of small capacities of the chemical manufactures was the characteristic of this period. The development of the basic chemical industry was reduced. But a fair part of export was based on the repro materials - raw materials and half-products; more than 85% of the basic chemical industry products were exported. Till the Second World War the Yugoslav chemical industry, because of a great number of circumstances, stayed as an undeveloped industrial branch, with a number of small economically poor firms with out-of-date technologies.

After the Second World War the development of the Yugoslav chemical industry came under particular conditions, specifying two periods: before and after the year 1956.

The main characteristic of the development in the earlier period, before 1956, was favouring of inorganic basic chemical industry, based quite or mostly on the domestic raw materials (the production of sulphuric acid, soda, chlorine, nitrogen compounds, phosphate fertilizers, calcium carbide and so on). The development of

the organic chemical industry in this period was not remarkable, with the exception of polyvinylchloride production.

In the second period, after 1956, the funds were invested in arising of the greater plants of organic and inorganic basic chemical industry, especially of that, based on the basis for petrochemistry. In the period from 1956 to 1958 a great number of factories was put in work, as shown in the table I.

Table I. More important capacities built after 1956

The Chemical-electro-metallurgy factory "Yugavten" Jegunovoi	55.000 t. of cyanamide 50.000 t. of calcium carbide, the various salts, ferrosiliciums and so on
The Factory of sulphuric acid Zos	234.000 t. of sulphuric acid
The Chemical Industry Konevsko Mitrovica	130.000 t. of sulphuric acid 250.000 t. of phosphate fertilizers
The Factory of Phosphate fertilizers in Prilevo	575.000 t. of phosphate fertilizers
The Chemical Industry in Pančevo	560.000 t. of calcium ammonium nitrate
Organic Chemical Industry "OKT" Legrad	20.000 t. of polyethylene 10.000 t. of styrene 6.500 t. of polystyrene 6.200 t. of phenole 4.200 t. of acetone and so on

Table I - contd.

Organic Chemical Industry "OKIS" Skopje	4.500 t. of polycrylic acid 4.500 t. of polyvinyl chloride 2.000 t. of polyvinyl acetate 6.500 t. of chlorine 7.200 t. of caustic soda 6.000 t. of dodecylbenzene 200 t. of lindane and so on
The Factory of Polyamide Fibers in the Complex of the Chemical Factory of Koste	1.200 t. of polyamide fibers
The Factory of Synthetic Fibers "Progress" Prilezen	2.000 t. of polyamide fibers

The capacities of some of these factories were soon increased. So the production of calcium ammonium nitrate in the Chemical industry Pančevo was increased for further 450.000 t. The capacities in the manufacture and in pharmaceutical industry were extended, too.

Table II. The production of the basic chemical industry

(100 t.)

	1938	1954	1958	1970
Sulphuric acid	23,2	472,3	539	747
Nitric acid	-	227,3	340	379
Phosphoric acid	-	-	13	132
Ammonia	-	128,3	212	347
Chlorine	0,2	30,2	38	42
Phosphate fertilizers	27,8	935	963	1.254
Nitrate fertilizers	43,3	423	522	1.532

Table II - contd.

	1953	1964	1970	1970
Viscose products	-	21	32	52
Synthetic fibers	-	47	101	110,5

In this period the development of the basic chemical industry was the slowest one, because of insufficient investments in basic organic chemical production, especially in petrochemistry. This insufficiency should be the particular source of many difficulties in the next period.

If we take the extent of the basic raw material components production (ethylene, propylene and butadiene among alkenes, and benzene, toluene and xylene among aromatics), as a measure of development of organic chemical industry in Yugoslavia, we can draw the conclusion that Yugoslav organic chemical industry is still at the initial level of development. This level is remarkably under the effective necessities of the established degree of the Yugoslav economy development.

The basic organic chemical industry of Yugoslavia has been started in the period of the relatively "closed economy" in conditions of limited possibilities for the following of the world development in this branch. So this industry in Yugoslavia considerably differs from the industry in other countries which dictate the relations in the world market, not only in the view of capacities but in technology characteristics, too.

As prerequisite for the further increasing of all the mentioned in the next period in Yugoslavia, the organic chemical industry in the first place has to be intensively developed on the basis of materials for petrochemistry.

Table III. The consumption forecast of some important chemical products

(000 t)

	consumption		
	1975	1980	1985
I. Inorganic products			
Sulphuric acid	1283	1300	1300
Ammonia in t N	540	550	755
Chlorine	103	147	201
Caustic soda	163	205	230
Calcinated soda	253	326	457
Phosphate fertilizers in t P ₂ O ₅	400	463	500
Nitrate fertilizers in t N	450	500	600
II. Organic materials			
PVC	140-160	170-230	230-300
Polyolefin	50-150	100-230	270-400
Polyisoprene	20-25	40-50	50-60
Another artificial materials	340-375	210-340	320-350
Fibres products	60	75	35
Polyamide fibers	10	15	20
Polyacrylonitrile fibers	15	20	30
Polyester fibers	25	25	35
Another fibers	2	5	3
Synthetic rubber:			
- SBR	50	55	45
- cis-polybutadiene	15	25	30
- butyle	5	10	25

III. PRODUCTION, DEVELOPMENT AND PERSPECTIVE OF THE PETROLEUM INDUSTRY

There was no petroleum production in Yugoslavia until 1939, so that its development belongs to the period after the war. Between 1945 and 1970 it was produced 25 million tones of petroleum and about 4,5 mil. m³ of the natural gas. The improvement of this production made the influence on the petroleum consumption as energy source in our country and it supported the development of some energotechnological and petrochemical industrial capacities.

Table IV. Production of petroleum and of the natural gas
between 1960 and 1970

Year	Production of petroleum		Production of the natural gas	
	000 t	index (1)	000 t	index (1)
1960	944	100	-	-
1963	2.063	218	213	100
1966	2.222	235	107	622
1967	2.374	251	107	753
1968	2.494	264	109	671
1969	2.699	235	103	1100
1970	2.834	302	106	1377
			977	1354

Index (1) : Index relating to 1960 = 100

Index (2) : Index relating to the previous year

As the table IV shows, the production of petroleum and the natural gas during the last ten years is constantly but unsatisfactorily increasing. Besides the constant production increases the domestic petroleum could not satisfy the need for consumption in the passed period, so that along with production the import of petroleum grew up.

Table V. The participation of domestic and the imported petroleum in the whole manufacture
(in 000 t)

Year	Import	Export	Disposable for manufac- ture	Participation of domestic petroleum in the manufac- ture in %
1960	435	-	1.263	75,1
1965	1.107	-	2.950	71,1
1966	2.202	324	4.132	57,2
1967	2.547	322	4.495	52,5
1968	2.635	292	4.931	50,5
1969	3.300	257	5.724	47,4
1970	4.442	150	7.100	46,0

(The year production is shown in the table above)

In perspective, unless some unexpected rich sources could be found out, we can surely expect the tendency for the continual decrease in the consumption of domestic petroleum. This tendency is shown in the table V. Some reserves of petroleum and the natural gas, discovered up to now, but not exhausted yet, - bilinear reserves - amounted to about 63 million tonnes of the crude petroleum and 40 mil. km³ of the natural gas at the beginning of 1970.

The consumption of the petroleum derivatives in Yugoslavia, although in remarkable growth during the last few years, is still low. As it can be seen from the table VI, the participation of the chemical industry in this consumption is continually increasing. The same remarkable increase in consumption of gasoline as a fuel and for the chemical industry's purposes is expected in the future.

The constant increase in consumption of all other petroleum derivatives as: liquefied gases, the special benzines, oils, the lubricants, bitumen, paraffins and so on is expected. The best consumer of these derivatives is industry with the participation of

about 60%. About 25% belongs to the general and individual consumptions. In perspective the industry is supposed to be the best consumer of those derivatives.

Table VI. Participation of particular sectors in the whole gasoline consumption

Sector of consumption	1960	1965	1966	(%) 1957
Industry	14,6	24,4	25,2	23,7
Traffic	63,5	60,7	62,2	65,0
Agriculture	5,8	2,5	2,5	2,1
Personal, general and another consumptions	16,1	12,5	12,1	11,2

The capacities for the petroleum manufacture in Yugoslavia are increased in particular after 1964. By building the new capacities and conducting the modern processes, the possibilities for the petroleum manufacture are increased and the quality of derivatives is improved: now they quite correspond to the world standards. Meanwhile, too fast including of the new capacities in production brought to the incomplete use of them, so that refineries worked with about 2/3 of installed capacities.

According to the available results obtained by the research works, we can expect the production of realization about $4,5 \times 10^6$ tonnes of the crude petroleum and about $2,5 \times 10^9$ m³ of the natural gas until 1975. The consumption of the petroleum derivatives will grow up, too; in 1975 it is expected to reach the level of about 12×10^6 t with the tendency of the further continuous increase. With regard to the available sources of domestic raw materials, the refinery capacities, as well as those in planning, will be sufficient through their complete use, until 1975. In the next five years period it will be necessary to build the new capacities for the petroleum manufacture necessary to ensure the continuity of supplying with petroleum derivatives after 1975.

IV. ORGANISATION AND ACTUAL PROBLEMS OF CATALYTIC RESEARCH IN YUGOSLAVIA

Although the problems of catalysis are present in the chemical industry of Yugoslavia about a century, and although the number of catalytic plants have been constructed in Yugoslavia in the period of scientific and technical development of catalysis in the world, the systematic investigation in this field has been organized only ten years ago. This was contributed by many circumstances concerning the industrial production, as well as the scientific investigations.

In the period between two wars, while the chemical industry of Yugoslavia was developing under the influence of the foreign capital, the scientific investigations were performed mainly abroad, while the experts employed in Yugoslav factories applied the obtained results in the production control.

In the first part of the afterwar development, which came after the long stagnation, there were the strong disproportion between the extension of the actual problems and the number of existing experts. Later on, when the number of experts in industry sensibly increased, the problems of catalysis did not get the real meaning for many reasons: in the new plants the license work conditions limited the competencies of the domestic experts and inhibited their interest for the problems of this kind; in the old plants the relatively small contribution of the catalyst price in the total expenses of production, left the wrong impression about the real economical aspect of catalysis problems. Besides all that, the experts employed in the chemical industry of Yugoslavia were not professionally high educated what was necessary for deeper insight in this problem.

In the University teaching, the catalysis was taught very fragmentary, and it was involved in other courses as general chemistry, physical chemistry, organic chemical technology etc. Involvement

of catalysis in the regular teaching started as a separate subject at the Faculty of Technology, University of Belgrade. Since 1950 catalysis with the laboratory practice is introduced as the regular subject at Chemical engineering at the Faculty of Technology, University of Novi Sad. The courses of theoretical and applied catalysis have been involved since a few years ago as the subjects of the postgraduate lectures at the Universities of Beograd, Sremske, Novi Sad.

Meanwhile, the lecture programs of these subjects are not in proportion with other lectures and they do not include all the important aspects of the theories and the applied catalysis.

The deficiency of the systematical school education in the field of catalysis could not be compensated adequately by education of the engineering experts taught the practice because of the permanent fluctuations of the study. The situation of the experts, very characteristic for the period of the intensive chemical industry development, was present in the Yugoslav industry, too. Although the structure of workers in the new built factories became even, less or less, during considerable period of time, the fluctuation of the workers, especially in the factories which are far from the cities, acts as a negative factor in relation to the necessary accumulation of the knowledge and the experience.

In the period of the interwar development of the scientific research in Yugoslavia, when the research groups and schools are organized in the special fields of chemistry, the problem of catalysts is investigated partially, on the level which was needed for solving the other problems. A few initiatives proceeded to the organization of the more complex systematical research of catalysts. In the Chemistry Department of the Faculty of Technology in Belgrade 1945-46 it was founded the Institute for Technical catalysts which ceased to work because of the personnel changes. In the Faculty of technology in Belgrade in fifties of this century, it was envisaged the problem of the engineering catalysis, but given the difficulties occurred not suitable for more permanent organization, catalyst

development of this technology.

Actually the development of the most complex specialized research in the field of catalysis in Yugoslavia started more than 10 years ago through the foundation of the Department for catalysis of the Institute for chemistry, technology and metallurgy in Beograd. This Department represents now not large but manifold equipped specialized institution, which initiates and coordinates all the important scientific-research projects in the field of catalysis, during the last 10 years.

Along with continual applied research which this Department performs, it is also concerned in the practical investigation and optimization investigations for industry.

Beside the Department for catalysis of the Institute for chemistry, technology and metallurgy, some other Institutions as the Chemical Institution Boris Kidrič, Ljubljana, Departments of the University of Beograd, the University of Sarajevo, the University of Split, the University of Novi Sad and the Research Institutes of some big enterprises as the Chemical Industry Pančevo, ZIM - Zagreb and so on are concerned with catalysis in a small degree. The main part of catalysis investigations in the industrial laboratories and for the necessity of industry at all, are performed in cooperation with the Department for catalysis of Institute for Chemistry, technology and metallurgy, so that in the further exposure you can find the investigations done in this Department.

Not talking about the fundamental problems of catalysis which are treated on the basis of the contract with the scientific funds, we are going to present the most important lines of the research development for the industrial purposes.

One of the first problems which drew attention of the industry, is the optimization of the kinetics as well as the catalyst choice. Now the catalyst for polyvinyl chloride, which is an exception, there is no catalyst produced in Yugoslavia, and they have to be imported. Ten years ago our firms were busy in selecting the opti-

lysts, trusting the renown foreign producers.

Nowdays they clearly show the tendency to take into consideration the professional criteris too, beside the commercial reasons, in deciding between the large number of offers.

The other problem which initiates the cooperation between the industry and the Institute is investigation of the influence of different process parameters on the activity and stability of catalysts. The motives of this interests are different; from the insufficiently stabilized process conditions in the older, not enough automatized plants, to the variations in the quality of the new materials and process interruptions because of the mechanical damages. The interest for this problem is inspired by the everyday practice after the garantee period and the cooperation with the scientific institute was supported by the arbitration investigations.

To the problem of the production of catalysts for the necessities of the chemical industry is not given too much attention until now, mainly because of the insufficiencies in the economical indices about the profitable production of the catalysts for the present capacities of the catalytic plants in Yugoslavia. The interest of several enterprises for making the common reserves of catalysts for the special processes is shown. The investigations, started in order to unify the process conditions and the catalysts, could be the first phase in making the domestic market ready for catalysts produced in the country.

Nowdays there is in industry the growing interest for organization of the permanent cooperation with the institute, concerning a broader research programme in the field of catalysis. In this way the necessary knowledge of the research staff people for unexpected intervention investigations would be ensured. The convenient circumstance in the organization of the permanent research in the field of catalysis is presented by the association of industry in order to form one common research center in the institute. The advantage of such a work is that it enables one research team to perform in-

vestigations from the fundamental ones to the directly applied according to the unique research approach, which ensure determination of the particular problems in all the phases of their investigations up further on.

Along to the development of the permanent research in the field of catalysis the interest for the permanent education of people working in the industry is evident, in the form of periodical seminars on actual problems of the theoretical and applied catalysis. We started with such seminars in 1970 in organization of the Department for Catalysis of the Institute for Chemistry, Technology and Metallurgy, Belgrade - with the participation of some eminent foreign lecturers. At the 1st Seminar, the lecturers were from the Institute for Catalysis in Novosibirsk and for the next seminars the lecturers from other well-known institutions are expected also.

7. ACTUAL PROBLEMS OF THE INVESTIGATIONS FOR THE INDUSTRIAL PURPOSES

Among the problems emphasized in the up-to-date scientific-technical collaboration of the Institute with industry, at this moment the attention is drawn to the standardization of methods for the examination of catalysts, the analysis of the applicability of data given for the characterization of the catalysts and the finding out the methods for the study of the catalyst's aging within reactors.

As a result of the variety of the catalytic systems, a large number of factors influencing their activity and variety of the process conditions in the particular plants, the problem of standardization in catalysis become more complex than in other fields. However, the practice showed that the comparison of different samples of catalysts is necessary. In order to establish rational basis

for such a comparison a tendency exists towards:

- selection of characteristic properties for particular systems;
- establishment of the correlations between various process conditions and the degree of varying of values of some characteristic of catalysts;
- definition of methods for particular characteristic evaluation.

Our enterprises know that certain research institutions and industrial plants throughout the world have their own standards, which are in disposal only partially.

As a result, a tendency for the establishing the own standards appears, which may be advantageous - to ensure a steadier work but also disadvantageous, as it can lead to the difficulties if accepted method give the different results. When for instance, the term "catalytic activity" is cited, usually it is not explained whether the activity based on the unity of area, volume or the weight of catalyst is understood nor by which method and under what conditions (dynamic, static) was determined. Or, concerning the data on the mechanical strength of catalyst, the terms: "strength", "crush strength" or "compressive side strength" appear, but no details on the methods of determination of the characteristics mentioned are given. As the results of the mechanical strength determination of the catalysts differ very widely and the results are largely influenced by the method used it would be very useful that giving the data, the method of determination is given also. Similar remarks bear upon other methods of investigation.

Along with the standardization of methods, a question of the applicability of data was risen. A number of data, given as characteristic of an catalyst, are not sufficient, if complementary data are not given, or, for certain types of catalysts some usual data are of no value. Good example is the vanadium catalyst for the production of sulphuric acid whose activity is influenced by the thickness of the liquid layer under process conditions. The data on the

magnitude of specific surface area are of limited value if the relationship between the active mass and carrier is not known. For other catalysts, the data on the percentage of the basic active component are insufficient if the role of other components in the mechanism of catalysis is not known and if their amount is not cited. The same approach is applied in recent programs for the investigations of the stationary phase composition of the catalysts under process conditions and the interaction between the catalysts and reaction media.

Our industry has a growing interest for the methods of studying the dynamics of the catalyst aging in reactor. This problem is of importance because of the very high costs of production breaks so that the planning of the catalyst's exchange along with the other interventions in the plant maintaining as well as an adaptation of the process conditions to the observed tendencies of aging, would be desirable. This question is very complex and its solving needs a deeper fundamental knowledge of the catalytic system, a certain engineering experience and the corresponding computer techniques. In lack of all of these elements, the services outside the country, where such analyses are possible are used. However, the interest for such investigations is emphasized and a collaboration in solving these problems is more than desirable.



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