PETROCHEMICAL RESEARCH AND DEVELOPMENT

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In April 1973, the Hungarian Government has passed a resolution in which the Petrochemical Central Development Program has been approved. This program is under the direction of the Ministry of Heavy Industries, and it is the youngest from among the central development programs. The programs approved by the Government aim at the shaping of a more efficient economic structure, but among these, the Petrochemical Central Development Program is of prominent importance since it involves not only the transformation of the production structure but it influences decisively the advance of chemical processing practically in all consumption fields. It helps fulfil other central development programs, *e. g.* the light structure building program, the vehicle program. It has a direct influence on the living standards of Hungary's each inhabitant since our country's population shares also immediately the consumption advantages of products manufactured within the framework of the petrochemical program.

The Petrochemical Central Development Program has started with the aim of satisfying about a three-quarters part of the petrochemical product demand showing a sevenfold increase from domestic production, and/or by economic integration product exchanges among the socialist countries.

Since the passing of the resolution, only three years elapsed but important results have been achieved already in the field of domestic petrochemical development. Tasks prescribed in the 4th Five-Year-Plan were fulfilled. The Leninváros Olefin Plant, Hungary's biggest petrochemical plant was put into operation on schedule which produces undisturbedly meeting the Soviet—Hungarian interstate agreement, supplying the Kalush Chemical and Metallurgical Combine with ethylene according to contract. In Tisza Chemical Combine, the high-pressure polyethylene production was almost doubled through plant intensification. By way of intensification, the PVC-capacity grew in Borsod Chemical Combine and the PAN synthetic fibre production started in the Viscose Plant. Domestic plastic processing capacity was doubled as compared to that in 1970 approaching the amount of 1/4 million ton per year. Domestic production of petrochemical aromatics is developed at the Danube

Refineries. In addition to benzene and toluene production, also the production of *o*-xylene has been started this year.

Since the resolution, hardly three years passed. Nevertheless, important results have been achieved that are due to measures taken in the course of careful and manysided preparation of the resolution permitting this development. From among these, the most significant one is the Hungarian—Soviet petrochemical agreement concluded in September 1970. Moreover, other bilateral petrochemical contracts were made within the framework of the socialist economic integration.

There are basic conditions for shaping a petrochemical industry, such as the general stage of development of the economy, a suitable energy structure, the level of the motorization, a suitable order of magnitude of the oil and gas industry of the country in question, etc.

The petrochemical activity cannot be separated from the above problems of a given country since the petrochemistry can be created as a direct concomitant of the petroleum processing stock flow in the course of the utilization of the petroleum as an energy bearer and as a chemical feedstock in a complex way. The present decade has created the possibility of developing domestic petrochemistry.

In Hungary's energy structure, the coal played the biggest role in 1970 with its 50 per cent share. In the seventies, the leading role was taken over by hydrocarbons. In 1970, the share of the hydrocarbons was 43 per cent and this share will rise up to 65—66 per cent till 1980. In addition to the increase of the share, the growth of the absolute amount of energy consumption requires a speedy augmentation of the hydrocarbon amount used.

Hungary's energy demand is about to rise to one and a half times as great in this time. Hydrocarbon demand was 9.5 million tons in 1970 and it approaches 25 milllion tons in 1980.

The speedy growth of the hydrocarbon demands outlined above is given reason by the fact that the seventies are marked by a breakthrough of motorization in Hungary. While the country's internal combustion engine stock hardly exceeded 20 million HP in 1970, this value is rising to nearly fourfold that is to 80 million HP in 1980. Within this, the passenger car park showed a fivefold growth.

The above reshaping of the energy structure, as well as the increase in engine fuel demand necessitate the prompt augmentation of petroleum processing capacities, *viz.* to twofold in the course of this decade. But even more important is the growth of verticality within the processing capacity, the expansion of secondary methods.

This is the background upon which the development of the petrochemistry is based in our decade. While in 1970 the consumption of petrochemicals hardly amounted to 2 per cent of the hydrocarbon consumption, about 7 to

8 per cent of the total petroleum and petroleum products will have been realized as a petrochemical feedstock by 1980 which can be compared with consumption rate of the countries having well developed petrochemical structure. Here, the ammonia production was not taken into account.

The above described industrial political decisions naturally involve an important hydrocarbon demand.

The problem of raw materials

Hungary, like other countries in Middle Europe, is highly dependent on import which she assures from the Soviet Union within the socialist economic integration. But our country's potentialities should be considered, too. The industry was given the task by the Government in the seventies to develop 60 million tons of commercial hydrocarbon reserves of which 32.4 million tons were developed in the first half of the decade. The scientific and technical activity in the field of the petroleum and gas exploration is an important factor of Hungary's hydrocarbon supply, especially as far as domestic gas reserves are concerned.

The most important petrochemical *basic compounds* and the manufacturing possibilities thereof are tabulated in Table I. If this Table is looked at from the view-point of *raw materials* then it can be seen that the petrochemistry primarily requires low molecular weight hydrocarbons as a raw material.

The production of the highest volume petrochemical basic compound (ethylene) might be started from ethane or, in want of this, from s.r. gasoline, in the most convenient way. According to the sense, the s.r. gasoline is the basic material for producing aromatics, too. Consequently, the petrochemical development is decisively depending on s.r. gasoline as a raw material. Figure 1 shows the main stock flows of the petroleum processing industry to be expected at the end of this decade through a Shankey diagram. Here, it can clearly be seen that domestic petrochemical development is making preparations for further processing the gasoline as a raw material. At the lower limit of the economic capacity, the olefine production requires 1 million ton gasoline per year as a raw material background and the production of aromatics of an order of magnitude of economic capacity can also be build at least on 1 million ton gasoline as raw material demand.

This gasoline amount can be assured if more than 10 million tons crude are processed. This value will be attained by our processing industry in this decade.

Not only quantity problems cause heavy problems in the field of petrochemical raw materials. The production of petrochemical basic compounds depends to a great extent on the composition of distillates of the crudes pro-

Table I

Significant petrochemical basic compounds and the production thereof

	Produced	
	from what?	how?
Paraffinic hydrocarbons		
ethane	natural gas, refinery gas	absorption + low-temperature distillation
propane	natural gas, refinery gas	absorption + distillation
	petroleum	distillation
isobutane	natural gas, refinery gas	absorption + distillation
	petroleum	distillation
	n-butane	isomerization
n-butane	natural gas, refinery gas	absorption + distillation
	petroleum	distillation
isopentane	petroleum	distillation
	n-pentane	isomerization
n-pentane	petroleum	distillation
liquid normal paraffins	gas oil	molecular sieve adsorption, urea adduction
Naphthenes		
cyclohexane	benzene	hydrogenation
Aromatics		
benzene	gasoline	reforming + extraction + distillation
	gasoline, gas oil	pyrolysis + hydrogenation + + extraction + distillation
	toluene	dealkylation + transalkylation
toluene	gasoline	reforming + extraction + distillation
ethyl benzene	reformed gasoline	extraction + distillation
	benzene ethylene	alkylation
paraxylene	reformed gasoline	extraction + distillation + crystallization (or adsorption)
	reformed gasoline	extraction + distillation + isomerization + crystallization (or adsorption)
orthoxylene	reformed gasoline	extraction + distillation
	reformed gasoline	extraction + distillation + isomerization
Olefines		
ethylene	ethane	pyrolysis + distillation
	gasoline, natural gas, petr.	pyrolysis + distillation
propylene	gasoline	pyrolysis + distillation
butylenes	gasoline	pyrolysis + distillation
C_{12} olefines	propylene	polymerization
	n-dodecane	dehydrogenation + adsorption
Acetylene		
acetylene	natural gas, petroleum distillates	partial oxidation + extraction
	natural gas, petroleum distillates	electric arc process + extraction
	natural gas, petroleum distillates	plasma cracking + extraction
Diolefines		
butadiene	gasoline	pyrolysis + distillation + extraction
isoprene	gasoline	pyrolysis + distillation + extraction
	isopentane	dehydrogenation
synthesis gas	natural gas	catalytic steam cracking

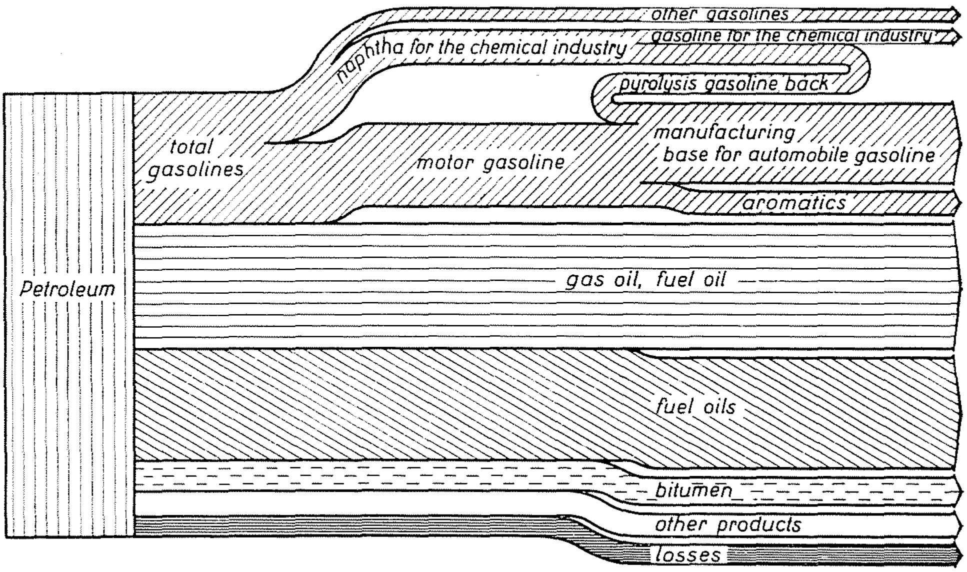


Fig. 1. Main stock flows of the petroleum processing industry.

cessed, on the proportion of various hydrocarbon types in the distillates which vary also in case of crudes coming from the same field.

Table II shows the gasoline quality of the crude produced from two horizons of the Romashkino oil field. In the Table it can be seen that the pro-

Table II

Hydrocarbon group composition of the gasoline fractions of Romashkino petroleum horizons

Dist. temp. ranges, C°	Yield (for petroleum)	d_4^{20}	Hydrocarbon content, %		
			aromatics	naphthenes	paraffins
Romashkino petroleum, Pashiysk horizon. Initial boiling point:					
60	4.1	0.6380	—	—	100
60— 95	4.4	0.7000	3	26	71
95—122	3.1	0.7346	8	27	65
122—150	4.6	0.7532	13	30	57
150—200	7.8	0.7791	19	31	50
Final boiling point: 200	24.0	0.7318	10	29	61
Romashkino petroleum, Uglenosniy horizon. Initial boiling point:					
62	2.2	0.6478	—	—	100
62— 95	2.6	0.6958	1	18	81
95—122	3.0	0.7203	2	23	75
122—150	3.4	0.7427	9	23	68
150—200	6.2	0.7800	14	27	59
Final boiling point: 200	17.4	0.7300	8	23	69

portion of the hydrocarbon fractions of the individual main producing horizons and, within this, also the proportion of the hydrocarbon types are significantly different. There is an even greater difference among the crudes coming from various trade sources.

When buying a process or a plant, the contractor specifies his terms according to raw material quality that is why one has to prepare to the impacts of raw material changes.

There are two ways to fulfil gasoline demand pertaining to the development of the petrochemistry. The one is the increase of amount of the petroleum processing, the other is the increase of the depth of the petroleum processing. By the latter it is meant that the white product, especially the gasoline yield of petroleum processing is increased by cat cracking. The application of the process allows the simultaneous fulfilment of the increasing motor gasoline demand and the petrochemical raw material demand, further the gasoline obtained by cat cracking supplies valuable high octane number components both to motor gasoline production and additional raw materials, *e. g.* the BB-fraction (butane-butylene) to the petrochemistry.

The white product yield of the domestic petroleum processing industry amounts at present to 51—52 per cent and, according to our plan, it is intended to increase it gradually to 59—60 per cent, in conformity with the development trends co-ordinated by the COMECON countries. The developed value does not reach the actual white product yield level of the USA but it exceeds that of Western Europe and it corresponds to the consumption demands to be expected. Of course, the possibilities of expanding the petrochemical basic material base should be examined further on.

The motor gasoline production and the petrochemical processing of gasoline is a reciprocative complex technology. The increasing octane number requirements and the increasing petroleum processing activity demand a more and more complex technology. This fact can be judged in Figs 2 and 3 showing a simplified flow diagram of our gasoline technology at the beginning and at the end of our decade. The production reactions of the most important petrochemical basic processes are shown in Table III. In this Table also the part of the catalytic processes employed in the petroleum processing industry can be seen which are directly connected to petrochemical processes, such as catalytic cracking, catalytic reforming and isomerization.

The Petrochemical Central Development Program realizes the selective industry developing principle. Domestic development objectives are shown in Fig. 4 in a synoptical picture of the development of the petrochemistry.

It appears from the data of the table that the reactions of highest volume are the heterogeneous catalytic reactions, the catalytic cracking, the catalytic reforming, the isomerization. The molecule breaking by pyrolysis is also of important volume. In the field of further processing petrochemical basic

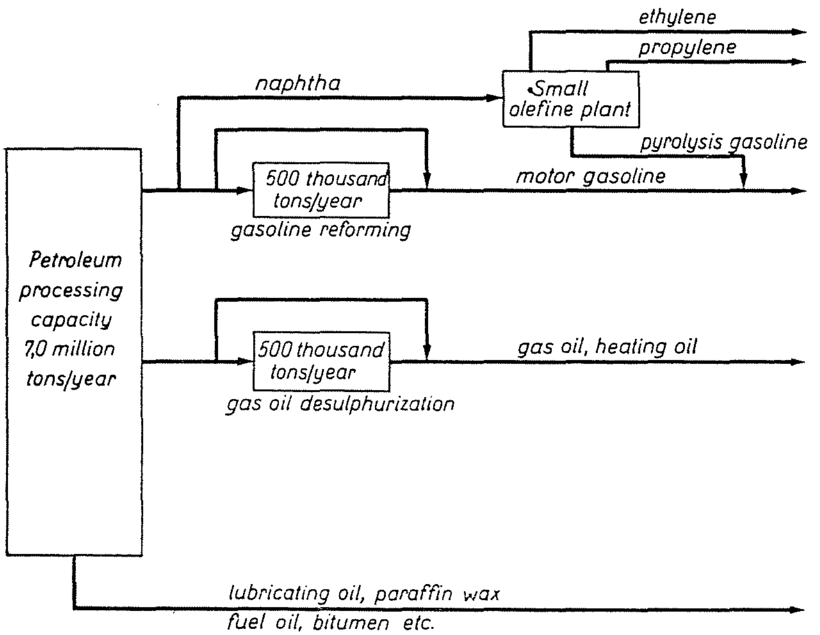


Fig. 2. Complex gasoline technology — 1970.

compounds there exists a plurality of processes, such as the various polymerization processes, the oxidation processes, the chlorination processes, further cracking of the products, such as the pyrolysis of dichlorethane, etc.

Table III

The most important domestic petrochemical basic processes

	Thermal	Heterogeneous catalytic
Pyrogenic processes	Naptha pyrolysis 940 thousand tons/year	Catalytic cracking 1000 — 1500 thousand tons/year
Reforming, isomerization		Gasoline reforming 1100 thousand tons/year Light gasoline isomerization 150 thousand tons/year
Oxidation processes		Maleic acid anhydride 12 thousand tons/year Phthalic acid anhydride 20 thousand tons/year

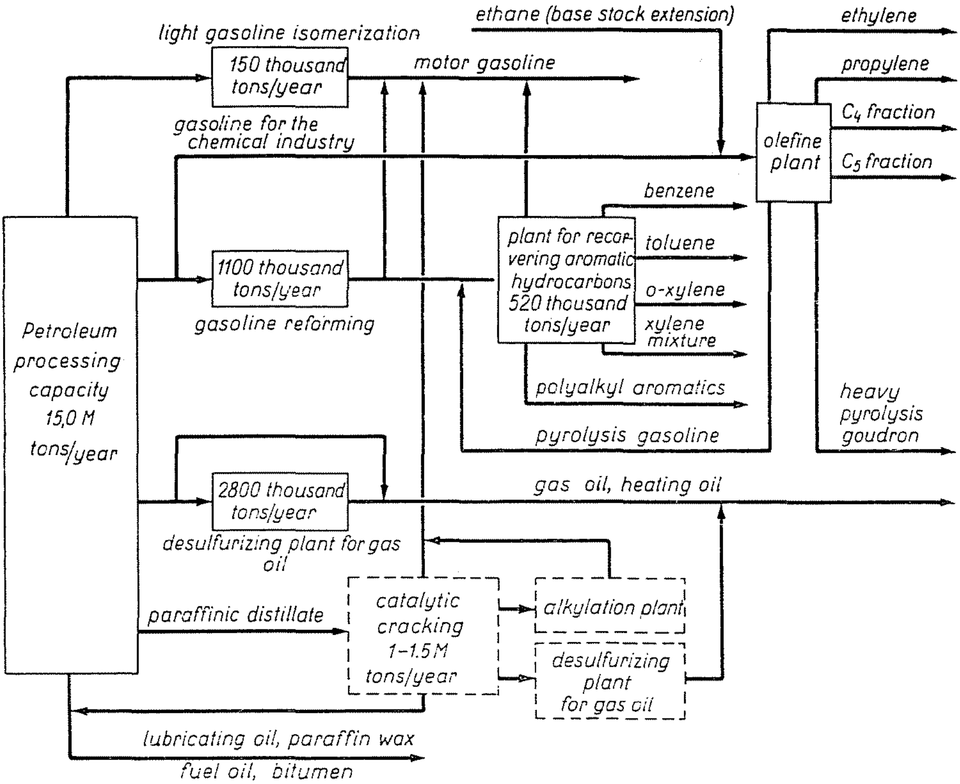
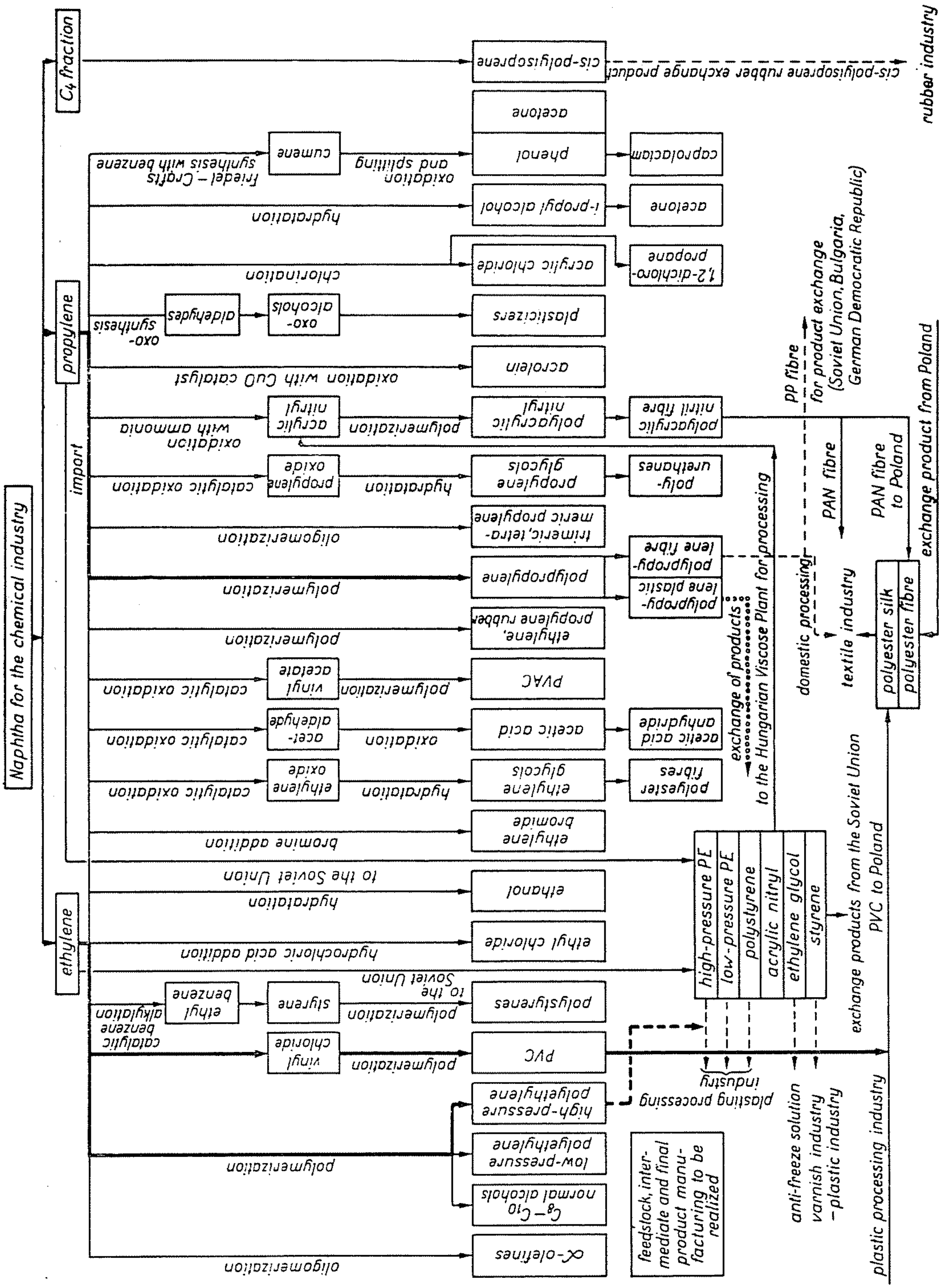


Fig. 3. Complex gasoline technology — 1980

Research and development

The role of scientific research in the development can be characterized, from the viewpoint of the choice of the project to be created, of the safeguarding of the technical level, by a general model. It is characteristic of this model that the development is always preceded by the research. The name of this activity, well-known in the international world, is: Research and Development (R + D). This general model, however, cannot be applied in case of decisions for selective industry development based upon international integration involving important structural changes. In this case, decisions are not originating from the results or domestic research results and are depending on macroeconomic considerations.

As it is well-known, the characteristic feature of these developments is that, in most cases, the basic methods and plant are bought fully prepared. This practice is proved all over the world and suitable in Hungary, too.


Naphtha for the chemical industry
C4 fraction
propylene
ethylene
import
cis-polyisoprene
cis-polyisoprene rubber exchange product
rubber industry
acetone
phenol
caprolactam
cumene
Friedel – Crafts synthesis with benzene
oxidation and splitting
i-propyl alcohol
hydratation
chlorination
acrylic chloride
1,2-dichloro-propane
oxo-synthesis
aldehydes
oxo-alcohols
plasticizers
oxidation with CuO catalyst
acrolein
oxidation with ammonia
acrylic nitryl
polymerization
polyacrylic nitryl
polyacrylic nitril fibre
catalytic oxidation
propylene oxide
propylene glycols
poly-urethanes
oligomerization
trimeric, tetra-meric propylene
polypropylene
polypropylene fibre
polypropylene plastic
ethylene, propylene rubber
vinyl acetate
PVAC
acet-aldehyde
oxidation
acetic acid
acetic acid anhydride
ethylene oxide
ethylene glycols
polyester fibres
bromine addition
ethylene bromide
to the Soviet Union
ethanol
hydrochloric acid addition
ethyl chloride
catalytic benzene alkylation
ethyl benzene
styrene
polystyrenes
vinyl chloride
PVC
high-pressure polyethylene
low-pressure polyethylene
C8 – C10 normal alcohols
α-olefines
high-pressure PE
low-pressure PE
polystyrene
acrylic nitryl
ethylene glycol
styrene
exchange products from the Soviet Union
PVC to Poland
plastic processing industry
plasting processing industry
anti-freeze solution
varnish industry – plastic industry
feedstock, inter-mediate and final product manu-facturing to be realized
exchange of products
to the Hungarian Viscose Plant for processing
domestic processing
textile industry
PP fibre for product exchange (Soviet Union, Bulgaria, German Democratic Republic)
PAN fibre
PAN fibre to Poland
exchange product from Poland
polyester silk
polyester fibre

The question has arisen whether the scientific research is needed at such development method. If experiences gathered from the whole world are analyzed, it should be stated that the scientific research is by all means required. There are only a few developing countries where the petroleum processing and the petrochemical industry has no scientific background. But these industry branches have become the properties of the nations by nationalization and a considerable amount of money is payed to the former multinational owners within the framework of contracts in order to assure a suitable scientific background. But significant efforts are being made also in these countries to create a domestic scientific background.

To the actual objectives of our petrochemical development, the original research and development model cannot be applied. Of course, the research tasks significantly change in this case. The first fundamental change is that the R + D sequence essentially alters, and the objectives of the R are determined by decisions relating to the D.

The fundamental industry development decisions are followed by the purchase of technological methods and equipment and the decision can be made only after the results of the preparatory researches.

The macro-economic decisions determine the basic material flow only. By using these technologies, numerous questions arise which become timely just with starting the production activity. Of these questions, the qualitative influence of the basic material on the process applied is mentioned. The optimization of the processes applied, the extension of running times, catalyst service life and regenerating questions, corrosion and environment protection questions, etc.

The decisions, however, leave a number of questions open. From among these, I first mention the question of further processing the so-called side stock flow of lower volume formed beside the decisive stock flows. In this case, a logical sequence of the original research and development model is required since the research results give possibilities for choosing the variants of the further processing. Research questions arisen in the flow diagram of the complex gasoline technology are shown in Fig. 5.

A typical example for the relation between the industry development based on central decisions and the domestic research activity is the research work pertinent to the C_4 fraction of the pyrolysis. The C_4 fraction of pyrolysis is exported in the course of the economic integration in return for the import of the synthetic rubber. No research works seem to be needed for further processing a product the whole amount of which is exported. But the fact is that the buyer of the C_4 fraction is interested in its butadiene content. Domestic research have shown that methyl-tertiary butyl ether can be produced from the isobutylene content of the fraction which is an efficient gasoline octane number improving additive. In the course of the process the butadiene con-

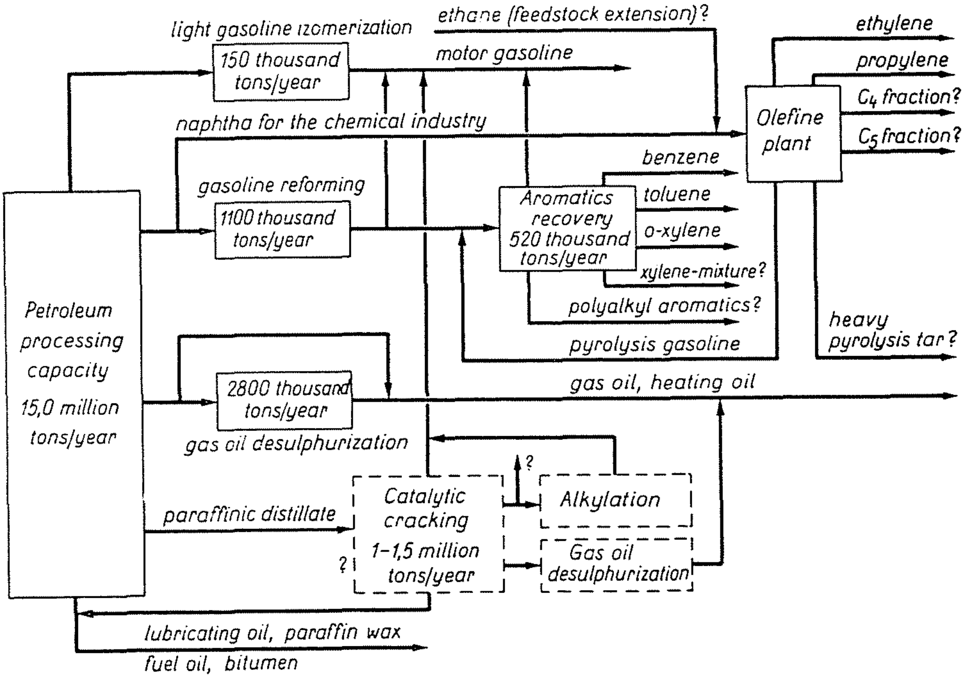


Fig. 5. Complex gasoline technology — 1980.

centration of the C_4 fraction increases which is, in addition to transport cost saving, an advantageous feature also from the viewpoint of butadiene recovery.

Problems of similar aspects may arise in connection with the C_5 fraction of the pyrolysis that goes for export in return for artificial rubber also within the framework of the economic integration.

There is a lot to investigate in the field of further processing the heavy pyrolysis goudron. The one way to be followed is perhaps the manufacture of petroleum coke which is of increasing importance because of electrode supply for our growing aluminium industry.

In association with pyrolysis, the problem of expanding the raw material should also be carefully analysed. Already earlier, rentability calculations were performed, based on research works, for the recovery of ethane from the Algyő natural gas. Although ethane is one of the most advantageous raw materials for producing ethylene, on the basis of our calculations, the processing of natural gases below 12 per cent ethane content to ethane was not economical.

Because of rising oil prices, the start of research has become timely again. But the research should cover the modification of the composition of base gasoline fraction for the chemical industry, and the importance of changing the hydrocarbon proportions, too.

One of the most significant field of domestic research objectives is the aromatics. The most expedient utilization order for the xylene stock should be found. The research covering so far the extraction of ortho- and para-xylene and the isomerization of meta-xylene should be enhanced and completed by research works pertinent to the oxidization of paraxylene into terephthalic acid.

It should be emphasized that a connection can be found in this field with an other big research program of the chemistry, *i. e.* with the domain of biologically active compounds.

The aromatics, and/or the intermediates originating from these are playing an important role when manufacturing pesticides. Exactly those aromatics that have a minor importance from the viewpoint of classical petrochemistry, such as toluene and meta-xylene are of primary importance in this context.

For instance, a method was published recently for making isophthalonitryl from metaxylene — similar to SOHIO process — which is an importante herbicide base stock.

The problem of polyalkyl aromatics should be clarified, too. Scientific researches going on in this field have already elucidated a number of questions in order to shape our objectives, *e.g.* in the question of mesithylene production; such a plant was put into operation in Italy recently and it is to be expected that the mesithylene question will be decided sooner or later.

Numerous problems have arisen in relation to the catalytic cracking plant that is able to supply base stocks not only for motor fuel production but also for the petrochemistry.

Application technological research for products is of very high importance. Its significance is supported by the fact that in the course of our decade the per capita synthetic material consumption will rise from 12 kg to 45 kg and the use of chemical fibres is growing in the textile industry of Hungary. Starting from the above consideration, the K-2 research target program, connecting to Petrochemical Central Development Program, will be elaborated. The Program Bureau for Target Program has been established and the determination of the research objectives detailed for the interested departments and institutions is beginning to take shape. The petrochemical research program is one of the biggest undertakings of Hungary's chemical research. It concerns the harmonized activity of scientific institutes, university chairs and company research departments belonging to 8 supreme authorities. The total number of these amounts to about 80. In addition to 3 institutes and 3 research groups of the Hungarian Academy of Sciences, 28 chairs of different universities have joined the research program.

Task of basic researches

The petrochemical methods have developed also in the first half of our decade. The chemical and reaction kinetic researches have revealed the importance of short contact times and, thus, the catalytic cracking process (riser technology) of high fluid yield and more recently the so-called millisecond pyrolysis (Kellog) were developed. The latter process has reached the maximum of ethylene yield obtainable by pipe-still pyrolysis which cannot be increased further theoretically. The results of chemical and kinetic examinations have made a new step of aromatizing processes possible by introducing the low- pressure reforming technology (CRR process).

These researches were performed simultaneously with a more precise knowledge of the regularities of the catalysis. In this way, the bimetallic catalysts have spread in the reforming, the molecular sieves in the destructive processes. In the polymerizations, the superactive catalysts requiring no subsequent washing-off have appeared. The latest industrial results rest upon a deeper knowledge of basic scientific laws, too.

Is basic research needed in Hungary? To this question only a unambiguous yes can be the reply. The presidium of the Hungarian Academy of Sciences has dealt with this problem already in 1973.

The importance of basic researches is very high. Catalytic reforming is going on in our country for a decade. The basic researches performed in this field have permitted a thorough analysis of the know-how supplying the process and the catalyst. These researches, by the application thereof in a proper direction, have made it possible for our plants to attain essentially more favourable results than those guaranteed by the process and catalyst suppliers and these research results contribute to the development of the technology also at the process suppliers. At the basic researches, however, scientific problems of trends to be expected for long-range advance have to be taken into account, such as influence of the extension of parameter limits, extremely high or extremely low temperatures, regularities arising in the high or low pressure ranges, etc.

The point is to what extent is the selectivity of scientific research motivated by the selective industry development. In the applied research it is doubtless that the effect of the selective industry development can be well appraised since the technologies realized in the country set up by all means higher claimse for research capacity and they are strongly bound to the equipment applied.

In the basic research, however, the selective basic research cannot be conceived but the advantages inherent in international co-operation should naturally be utilized in basic research, too. Preparations should be made for solving the possible problems, such as disproportionation of olefines, transalkylation of aromatics, oligomerization, etc.

The basic scientific research results can comprehensively be utilized. Oxidation researches are not discussed in this paper. These researches dealing with the oxidation of aromatics, in addition to attaining the aims set originally, have yielded the result that the new discoveries, completed by further researches, may help pave the way for a tertiary oil-recovery process, *i. e.* the underground partial oxidation method.

The further development of recognitions pertinent to surface phenomena related to heterogeneous catalysts may yield results also in the science of petroleum production.

The petrochemical research target program sets the following claims to basic researches:

1. Examination of petrochemical raw stocks and products.
2. Analytical material testing, methodical researches for hydrocarbons and direct hydrocarbon derivates.
3. Examination of hydrocarbon chemical reactions inherent in the transformation of various bonds.
4. Investigation of the thermal and catalytic transformation reactions and technologies for hydrocarbons as well as of chemical utilization of reaction products.
5. Examinations and investigations related to hydrocarbon chemistry catalysts.
6. Development of examination and measuring methods.
7. Soft-ware researches for elucidating hydrocarbon chemical reaction mechanisms.

Petrochemical basic research activity has been going on for a long time at research institutions dealing with petrochemistry, among others also at the institutes of the Hungarian Academy of Sciences. These works, as hydrocarbon chemical researches, were emphasized on department level. The sphere of the above co-ordination, however, should be extended by polymeric chemical researches, and it is expedient to rise the enlarged petrochemical researches developed like this to portfolio, secretary-general level. By doing so, a better joining of the researches to the Petrochemical Central Research Target Program would be promoted.

It would be suitable if the Academy of Sciences played its adequate coordinating, guiding role in basic research activities going on in some other fields, primarily at the university chairs.

This organization itself will yield no success alone. The efficiency of the researches would be enhanced by shaping a closer relationship among those dealing with petrochemical development and research problems, by recognizing each other's problems, by a closer co-operation with the petrochemical vertical manufacturing plants.

Another condition of the more efficient research is, in addition to the scientifically educated research staff, the safeguarding of an up-to-date instrumentation. The research background of newly set up petrochemical plants producing on a world level can be assured only by a large number of reliable informations. For this purpose, conventional research instrument base and methods are not appropriate.

The objectives of the research will be growing from year to year since the demands will increase for creating an intermediary base, for acquiring knowledges preparing the future, for the mastery of the petrochemistry, this important chapter of the modern chemical science, for educating the future chemist generation to perform these tasks, and for further training it.

The more developed the petrochemical industry becomes, the more the problems pertinent to the increasing verticality multiply and the more need arises for specialists dealing with research, planning and development to join their efforts for solving the tasks ahead.

Summary

A short review is given on the petrochemical developments achieved in Hungary since the approval of the Petrochemical Central Development Program in 1973. The olefin plant in Leninváros went on — stream on schedule, and the domestic production of petrochemical aromatics has developed. The demand for crude oil, for motor gasoline and petrochemical naphtha is discussed. In connection with petrochemical development, the link between (R)esearch and (D)evelopment is shortly analyzed. The earlier R+D sequence has essentially changed, and now the decisions made on the field of D determine the goals of R. In the development of petrochemical processes, basic research has a very important role and even smaller economic units like Hungary cannot be without basic research. The major tasks of basic petrochemical research in Hungary are given.

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