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Universal Plasma-chemical Module for Carbon-containing Raw Materials Treatment

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Abstract

A universal plasma-chemical module (PChM) for the industrial processing of different hydrocarbon raw material pyrolysis was designed and tested. Laboratory investigations for the plasma-chemical method of acetylene production from natural gas and different coals were made. Similar laboratory tests on the industrial production of acetylene as a raw material for organic synthesis were developed using the PChM. A comparison of the suggested plasma-chemical method with the traditional process of acetylene production were carried out. The outlook of the plasma-chemical method was shown.

Key words : plasma torch, plasma-chemical reactor, carbon-containing raw, pyrolysis.

1. Introduction

1.1. Plasma-chemical Reactors

The wide spread use of plasma torches in chemical and metallurgical industries, for testing heat shielding coatings of space modules, in aerodynamic and other studies on advanced problems such as the design of high power electric arc reactors with a long life time, uniform cross-section temperature and velocity fields, as well as high heat and electrical efficiencies.[1,2]

The temperature fields of a plasma stream in the region of the plasma torch exit nozzle are usually non-uniform, particularly for a plasma torch with a self-stabilizing arc length.[3] Fluctuations of power, flow temperature and pressure may be in the vicinity of tens of percentage points. A mixing chamber needs to be inserted in the plasma-chemical installation to improve the kinematic and dynamic characteristics of the reactor.

Therefore, an effective mixing chamber is essential.

Technological installation power has been increasing with the increasing use of electric arc gas heaters. The power of one plasma torch unit exceed tens of MW.[46] However, the lifespan of these arc heaters is very low. This is, because of the high arc currents that are normally used. The use of a mixing chamber with a small number of plasma torches makes it possible to solve the problem of designing a plasma-chemical reactor with the required power, uniform temperature, pressure, velocity fields and long lifetime.

Parts of working gas are injected into the mixing chamber of a technological reactor through the plasma torches at high temperature. Other parts (i.e. a single gas, gas mixture, gas-powder mixture etc.) inject through the feeding system in the reactor directly. Plasma-technological processes are generally carried out in gas streams of Ar, H_2, N_2, air, natural gas, steam, and other gases and mixtures.[7] Those gases have quite different characteristics. Therefore, these characteristics need to be taken into account. The characteristics include the possibility of reaching a high enthalpy, the use of the working gas as one of the reagents, passivity to final products.

A high importance at the same time with a choice of the optimal technological parameters of the reactor in terms of the engineering problems associated its design is required. This reactor is obliged to provide:

- feeding of raw materials in the most heated region of the gas stream for the maximum treatment;
- high stability of the working parameters in the reaction zone; and
- exclusion (if required) oxidation of the reaction products in the case of installation decompression.

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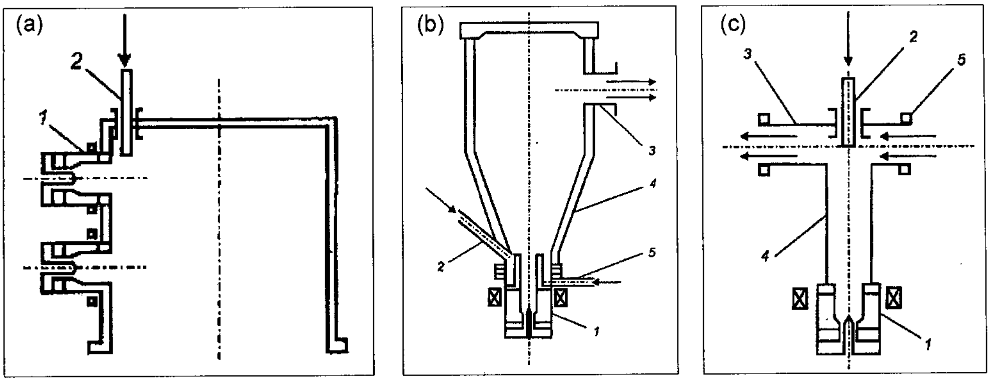


(a) with parallel jets (b) with a fluidized bed (c) with an anti-parallel raw feed.
1-plasma torch, 2-raw feed, 3-off gases outlet, 4-reactor body, 5-cold gases inject.

Fig. 1. The diagrams of reactors.

These problems and of the need to reduce the size of the plasma equipment stipulated for the development of a number of plasma reactor designs. A review of russian developments in this area is given in the monography.[8] It should also be noted that only the reactor designs, which are intended for different variants of stream processes (except of one-jet co-current flow reactor, which will be discussed further). Those reactor diagrams are given in Fig. 1. Three main schemes of the reaction chamber:

a) a reactor with parallel jets. In this reactor a few plasma torches are disposed in one plane, which ensures the effective treatment of polydispersive raw materials;
b) a reactor with a fluidized bed. In this reactor, raw materials arrive in the high- temperature area of the plasma jet from surrounding fluidized bed zone;
c) a reactor with an aligned attached plasma torch including anti-parallel raw feeding.

Another scheme is a multi-arc reactor, where several plasma jets are inject the raw material stream, which allows the possibility of designing a high power plasma apparatus.

Therefore, the reactors for the plasma-stream treatment of raw materials may be used in the technological circuits of industrial production, as they are currently used in chemical technologies. However, the main question that applied plasma-chemistry does not solve, is the development of a scientific basis of plasma-chemical reactor design. Generally, the reactor scheme and its design are carried out by empirical way.[1)]

This conclusion can be confirmed by some known elaborations, which connected with municipal and industrial waste treatment.[9-15)] Some variants of the plasma-chemical installations suggested in these articles are based on the experience of furnace burning or the treatment of waste or raw materials. Part of this work is to determine a more effective way of using plasma-chemical reactor treatment. These types of installations are used in the production and treatment of oxides, nitrides, borides of metals powders, in producing ultra-fine powders, and in industrial production of TiO_2, MgO and other powders. An example of the large-scale use of a universal plasma-chemical unit for treating natural gas and coal is presented in this article.

2. Experimental Set

2.1. Universal Plasma-chemical Module

A great deal of attention has paid to the treatment processes of carbon-containing compositions in plasma-chemical reactors. Despite the large number of compositions, the treatment methods are based on their destruction in a high-temperature plasma stream. These include the high-temperature pyrolysis of carbon-containing compositions in a restoring hydrogen plasma, and the incomplete oxidation of carbon in a water-steam plasma, etc.[16-19)]

The main element of this high-temperature destruction

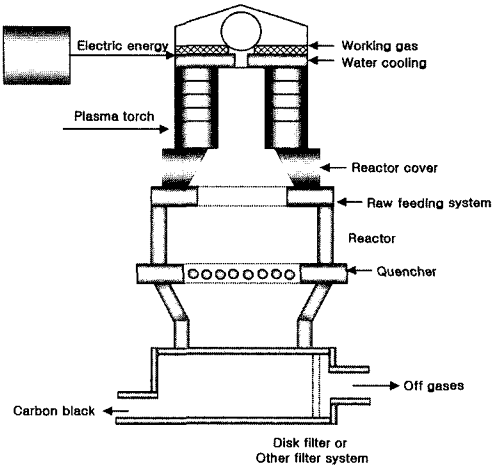


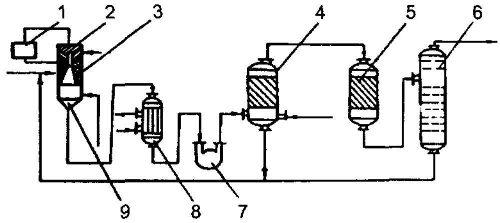
Fig. 2. Universal plasma-chemical module.

installation is the plasma-chemical reactor unit. In particular, a universal concurrent flow plasma-chemical module was designed and produced in serial by group of Russian enterprises for the industrial treatment of hydrocarbons, carbon-containing waste, coal and other raw materials. They include the joint-stock scientific-industrial companies "Technolog" (Sterlitamak, Bashkiria), Institutes of Thermophysics, Theoretical and Applied Mechanics SB RAS and a joint-stock company "NOVOSIBIRSK NIIChimMash" (Novosibirsk). In Russia, studies on this system were carried out on the base of the "CAUSTIC" joint-stock company (Sterlitamak).[16-18] There were two variants of this unit, the PChM-375 and PChM-750 (PChM-plasma-chemical module). Their capacities were 375 kg/h and 750 kg/h of raw materials, respectively.

A diagram of this unit is given in Fig. 2.

The main elements of the plasma-chemical module are:

- an electric-arc plasma torch in heating the working gas up to 35 thousand degrees. The power of the plasma torch is 750 kW for PChM-375 unit and 1,500 kW- for PChM- 750 unit. The plasma torch is supplied from a power supply - DC controlling rectifier,
- the plasma-chemical reactor that treats the raw mixes with a plasma stream and the main reactions occur,
- a quenching device that, allows the reaction products to cool fast and to stop the process at the moment of the best output of the desired product,
- a system of cleaning the reaction products from the solid admixtures, and
- a system for separating the desired products.



.1) power supply 2) plasma torch 3) reactor 4) selective purification 5) synthesis reactor 6) separation column 7) compressor 8) heat exchanger 9) quenching unit

Fig. 3. Technological set-up.

Depending on the initial and final products of the plasma-chemical treatment, it is possible to include other elements into the scheme. These include different raw feeding systems, a dilution system for the raw paste, different variants of the quenching systems, a disk filter for carbon black separation in the case of a raw gas raw, a filter system for separating solid particles and ash (cyclone, bag filter and others), systems for neutralizing toxic and hazardous products, and the utilization of the reaction by-products, etc.

A typical diagram of the use of a plasma-chemical unit in a technological set-up[17,18] is given in Fig. 3.

The plasma-chemical unit works in the following way. The working gas, such as hydrogen, is heated in the plasma torch, 2, by means of an electric arc up to 3,000 5,000 K. This gas (low-temperature plasma) is injected into the plasma-chemical reactor, 3, where it mixes with the raw materials. The raw materials are heated, evaporated and pyrolyzed to produce, for example, acetylene, hydrogen chloride, methane, and hydrogen. The pyrolysis gases are exposing to the high speed cooling by quenching system, 9, and supplementary cooling in a heat exchanger, 8. The cooling gas is compressed by a compressor, 7, and injected into the selective purification reactor, 4, where pyrolysis gases are separated from the undesirable admixtures. These admixtures are returned to the pyrolysis stage. Pure pyrolysis gases are guided to the synthesis reactor, 5, and after synthesis - to separation column, 6, where the main production of the synthesis separated. The waste from the separation column, 6, is returned as a raw material to the pyrolysis reactor, 3.

This universal plasma-chemical module has a wide range of possibilities in terms of work and the ability to adapt to many installations. Plasma torches with different powers can be used as the heat source. In real conditions of an industrial plant, plasma torches have been with powers up to 3 MW. Three or more plasma torches may be placed on the cover of the reactor. Therefore, the plasma system can reach a very high power. A working gas may be used practically in every gases and their mixtures. It is possible to control the time required for raw treatment over wide range of limits by changing the inside geometry and the length of the reactor. The temperature of the heat carrier may be changed over very wide range of limits. When the temperature in the reactor is T=1,000 1,500°C, the decomposition of all organic combinations occurs. In hydrogen plasma, unsaturated hydrocarbons are formed and in a water steam plasma synthesis-gas is generated. If instead of hydrogen, air (oxygen) is used as the heat carrier, the process of destruction of the complete compositions is accompanied with the oxidation of the formed compositions of the chemical elements. In such a way it is possible to destroy toxic substances e.g. pesticides and herbicides, bad herbicides and their mixtures, etc completely. On the final stage of the process the neutralization block is connected to the installation. It neutralizes components such as the oxides of sulfur, lead, phosphorus, etc. Installations for pyrolysis or the decomposition of toxic wastes are now at work in some chemical plants in Russia. Serial plasma-chemical units with a power of 750 and 1,500 kW are currently used.

3. Experimental Results and Their Discussion

3.1. Synthesis of Chemical Products from Natural Gas By Using Of Universal Plasma-chemical Module

Acetylene is made traditionally from natural gas by the way of thermal-oxidizing pyrolysis. Synthesis-gas ($CO+H_2$) is by-product as well as carbon black.[16]

The thermal-oxidizing pyrolysis of natural gas is joined to a considerable specific raw rate for acetylene synthesis (because of combustion). Only 23-25% of the methane is rated for acetylene synthesis. Its main part (55%) is used to maintain the high process temperature. Despite the fact that a considerable amount of synthesis-gas is generated (up to 10,000 nm^3 of mixture $CO+H_2$ per 1t of C_2H_2), the total energy consumption including the energy consumptions for oxygen production is 4.70 t of conventional fuel per 1 t of C_2H_2.

The physical heat of the reaction products is not used and technical carbon (carbon black) is not formed as a trade product.

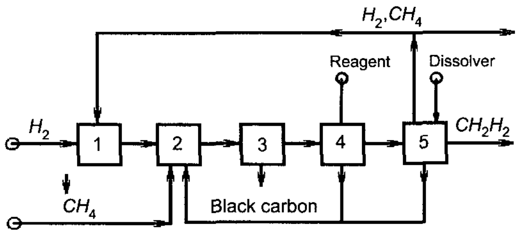
The scheme of gas cleaning from the admixtures is cumbersome and power-consuming. When organic dissolvers are used at the stage of acetylene formation, organic wastes are generated, which may not be used profitably.

The method of acetylene production, which is elaborated by the aforementioned enterprises concern plasma-chemical methods, which have been developed over the last 30 years.[20] The method is analogous to the method for the electro-cracking methane, which was elaborated by the "Hüls" company (Germany).[21] "Hüls" used natural gas as a raw materials, passing oil gases, gases from oil treatment, oil-chemistry waste etc. Similar processes were reported by the "Dupon de-Nemur" company. This method is sufficient universally, but it has some demerits. These include, carbon deposition inside the plasma-chemical reactor, and a high non-uniformity of the temperature fields in the reaction zone.

The method of pyrolyzing the hydrocarbon raw materials in a hydrogen plasma stream is deprived of these demerits.[16-18] The main elements of the installation of acetylene from natural gas (see Fig. 4.) are:

- the unit of raw material (natural gas) preparation;
- a universal plasma-chemical module, which includes an electric arc heater with a power supply and control system, a plasma-chemical reactor and a quenching system;
- a pyrolysis gas cleaning and cooling system;
- a pyrolysis gas compression and drying system;
- an acetylene separation system.

The pyrolysis of natural gas in a hydrogen plasma (Fig. 4) allows the raw input to be reduced; oxygen is not used for the process. The process demands hydrogen, which is used only at the plasma torch; in working regime one uses technical hydrogen (a hydrogen-methane fraction) after the stage of acetylene separation from the pyrolysis gases. Dry quenching of the pyrolysis gases in a special heat exchanger allows the use of physical heat of gases for water steam obtaining. Carbon



1) gas preparation 2) pyrolysis 3) black carbon separation 4) cleaning from homologues 5) acetylene separation.

Fig. 4. A schematic of acetylene production from natural gas using the plasma-chemical method.

black is separated from the gases in special filter as trade product. The gases are cleaned from the acetylene homologues in a one-stage process. The homologues, which are chemically joined to harmless organic compositions, are returned to the pyrolysis stage as a secondary raw feed. The exhausted dissolver also returns from the stage of acetylene separation to the pyrolysis stage. Excessive amounts of technical hydrogen may be used as both a fuel gas and in the processes of oil treatment.

In Table 1, there is a comparison of the technical-economical indices of the different ways of converting natural gas into acetylene. Compared to the thermal-oxidization process, plasma-chemical pyrolysis has the following advantages: a lower raw rate and a lower total energy consumption. Plasma-chemical treatment of natural gas provides wasteless technology for producing of acetylene from natural gas.

Tests of the processes of natural gas pyrolysis were carried out in a laboratory and pilot-plant installations. In the case of a pilot-plant installation using the PChM-750 module, the following results were obtained: raw material flow rate -200~250 nm^3/h; fractional conversion -65~68 mole%; volume concentration of C_2H_2 in gases -12~14%; electric energy expenditure -10 kWh per 1 kg of C_2H_2; total yield of C_2H_2 -85~95 kg/h. These results do not give the same yield as the best plasma-chemical processes of C_2H_2 production. As far as energy expenditure is concerned, all the acetylene production processes are similar -10~12 kWh per 1 kg C_2H_2.

3.2. Production of Acetylene from Coal

Coal is not only an energy-carrier but also a raw for the chemical industry. The coke-chemical industry is the most developed one.

Synthesis-gas (CO+H_2) is made from coal by conversion (at the sacrifice in oxidizers use - oxygen, air, and water steam). The large-tonnage production of methanol and other chemical products (including hydrocarbons according to the Fisher-Tropsh method)

Table 1. Comparison of the technical-economical indices of the different ways of converting natural gas into acetylene (per 1 t of C_2H_2).

Indices	unit of measure	Thermal-oxidizing process	Plasma-chemical process
Raw rate			
Natural gas	thousand of m^3	7.2	2.4
Energy consumptions			
Electricity	thousand of kWh	2.3	11.0
Steam	Gcal	5.0	0.2
Circulating water	thousand of m^3	0.9	0.35
Oxygen 95%	thousand of m^3	3.8	–
Dissolvers	kg	7.5	7.5
Products			
Synthesis-gas	thousand of m^3	10.2	–
Technical hydrogen	thousand of m^3	–	1.1
Total energy consumptions	t of equivalent fuel	4.7	3.6

Table 2. Comparison of the results of the different plasma-chemical methods for producing acetylene from coal production.

Indices	"Technolog" (Russia)	"AVCO" (USA)	"Hüls" (Germany)	
			1	2
Plasma torch power, kW	30	1,000	30	30
Productivity by coal, kg/h	up to 15	325	2.5~3.0	2.5~3.7
Electric energy consumption for coal pyrolysis, kWh/kg	2.8	3.1	3.4	10~12
Carbon gasification degree, %	80~85	no data	65	65
Electric energy consumption for acetylene production, kWh/kg	8.0	8.8	13.6	25~30
Volume concentration of C_2H_2 in pyrolysis gases, %	13.2	15.6	14~16	14~16
Total yield of C_2H_2 from coal, weight %	35	35	25	40

is based on synthesis-gas. Acetylene is made from coal through calcium carbide.

Acetylene production from coal via calcium carbide is labor consuming. Up to 30% of the initial coal is lost as a carbon oxide despite improvements in carbide furnaces technology (such as closed furnaces, use of furnace gases). This process is energy consuming - electricity consumption runs up to 12 kWh per kilograms of acetylene. Carbide technology is related to environment pollution. It generates waste-water (30 m^3 per 1 t of acetylene), and pollutes the atmosphere with dust and carbon oxides.

Plasma-chemical technology is considered to be intensive resource-saving one at the process of directly producing acetylene from coal.

Enterprises from many countries have carried out scientific - research and experimental - design works for acetylene production from coal by pyrolysis in a hydrogen plasma ("AVCO"(USA), "Hüls" (Germany)).[21-24]

As specialists from AVCOhave calculated, the acetylene produced by plasma-chemical coal pyrolysis is able to compete economically with ethylene from petroleum in the production of vinyl chloride and vinyl acetate. The results of technical-economical calculations from Germany have shown the superior economy of a plasma-chemical method over both the carbide method and thermal-oxidizing pyrolysis of methane.

Experimental research on the process of acetylene production by the plasma-chemical method was carried out using different coals from Russia. This work used samples of brown coal from Bashkiria, black coal from Kazakhstan and East Siberia[18,25] as the raw materials.

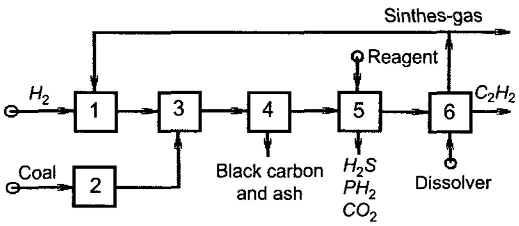
The study was carried out on an experimental installation with a power up to 30 kW. The installation consisted of a power supply, a plasma torch, a reactor, a weighing apparatus and a feeder of milled coal, a quenching device, a cyclone filter and a filter for separating the solid waste, and units for the plasma-forming gas supply and the cleaning system for the gases. The coal rate changed from 3 to 15 kg/hour. The plasma-forming gases were hydrogen, and hydrogen-containing gases. The coal from various fractions was charged into reactor.

The results of a comparison of acetylene production from coal by plasma-chemical methods are given in Table 2. The table also includes the results reported by "AVCO", H ls companies and the Sterlitamak Technolog company. The Russian method is more productive, consumes less power at a similar production output.

Taking these positive results into account, experimental works were carried out on a pilot-plant (on plasma-chemical installation with power up to 1,500 kW). The installation consisted of the following units: a unit for preparing the plasma-forming gas, coal, plasma-chemical pyrolysis unit, a unit to separate the solid waste, a unit to separate the admixtures, a unit to separate the acetylene(Fig. 5).

The gas composition was analyzed by gas chromatography. The gaseous products of the reaction consist of hydrogen, methane, acetylene, ethylene, carbon monoxide, carbon dioxide. The pyrolysis gases contained small amounts of hydrogen sulphite, phosphor hydrogen, acetylene homologues.

The effects of the specific energy requirements, the



1.-gas preparation 2.-coal preparation 3.-pyrolysis 4.-black carbon and ash separation 5.-cleaning from admixtures 6.-acetylene separation with dissolver.

Fig. 5. Block-scheme of acetylene production from coal using the plasma-chemical method.

coal particles, and the reactor design on the indices of the pyrolysis process were studied.

The maximum output of the acetylene was obtained from the brown coal samples -25% (for dry milled coal). The electricity consumptions for 1kg of acetylene production under the optimum conditions are 8 kWh during the pyrolysis stage.

Overall, the electricity consumption was approximately 10 kWh/kg acetylene. It was lower or close to the demands of the carbide method (near 11 kWh/kg). One plasma-chemical unit with a power of 1 MW is able to produce 700-800 t of acetylene per year. This process is a one-stage and practically does not pollute the atmosphere. Therefore, it has a great advantage from ecological point of view. In addition, the use of many plasma-chemical units is possible at a maximum; 1-2 of them are in reserve and the rest are working.

Table 3. gives a comparison of acetylene production from various other methods (carbide and plasma-chemical ones).

As a result of these experiments, the initial data for the engineering and economic calculations as well as the design of a large-scale plant for vinyl-chloride production from coal were collected. The main technical indexes of the process of plasma-chemical coal pyrolysis are:

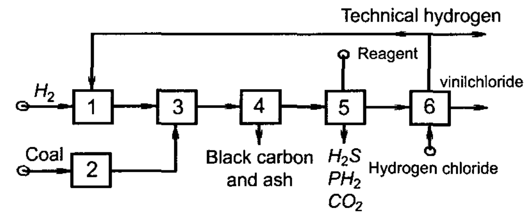
- carbon gasification degree -85%;
- acetylene output from coal -35%;
- electric energy expenditure for coal pyrolysis -3 kWh/kg.

The technological process of vinyl-chloride production from coal by plasma-chemical method includes the following stages (Fig. 6.):

1. Preparation of coal.
2. Preparation of the plasma-forming gas.
3. Coal pyrolysis.
4. Separation of solid waste.
5. Cleaning from admixtures (H_2S, PH_3, CO_2).
6. Synthesis of vinyl-chloride from diluted acety-lene.

Table 3. Comparison of the indices of processes of acetylene production from coal (consumption for 1 t of C_2H_2).

Indices		Unit of measurement	Carbide method	Plasma chemical method
Raw	Coke or black coal	t	3.6	–
	Brown coal	t	–	4.0
	Lime-stone	t	5.1	–
	Hydrogen	thousands of m^3	–	0.1
	Dissolver	kg	–	1.5
Energy resources	Electricity	thousands of kWh	11.5	10.0
	Water steam	t	0.3	–
	Chemically cleaned water	m^3	4	4
	Circulating water for cooling	m^3	320	250
By products	Furnace gas	thousands of m^3	1.20	–
	Synthesis-gas ($CO+H_2+CH_4$)	thousands of m^3	–	3.0
	Carbonic waste	t	–	1.2
Secondary energy resources	Water steam	t	–	4
	Total energy consumptions	t of equiv. fuel	4.3	3.5



1) gas preparation 2) coal preparation 3) pyrolysis 4) black carbon and ash separation 5) cleaning from admixtures 6) vinyl-chloride synthesis.

Fig. 6. Block-scheme of the production of vinyl-chloride from coal using the plasma-chemical method.

For the process realization both brown coal and hydrogen are needed. While combining the production in a chemical plant involving the electrolysis of common salt, the production is provided by electrolytic hydrogen in the start time and also by hydrogen chloride, which is synthesized from the electrolytic hydrogen and chlorine.

A part of the synthesis-gas is used (it is separated on the stage of vinyl-chloride synthesis) for continuous work on the pyrolysis stage. A small amount of synthesis-gas (CO, H_2) may be used as a fuel or after special cleaning as a raw material for producing methanol or hydrocarbons according the Fisher-Tropsh method. The use of synthesis-gas as a fuel in gas-turbine power plants makes it is possible to produce electricity for the plant's own needs. In this plant, of 3 MW of electricity will be produced in the case of an acetylene plant with of capacity of 1 ton per hour.

Eventually, the total energy consumption using the plasma-chemical method of acetylene production from coal are 1.2 times less than using carbide method.

The synthesis of vinyl-acetate also may be realized from diluted acetylene (i.e. without separating the acetylene from pyrolysis gases).

In such a way, the synthesis of vinyl chloride and vinyl acetate from diluted acetylene makes the technological scheme of its production easier, and results in reduction of the operation costs.

4. Conclusions

1. A universal plasma-chemical module for the industrial process of the plasma-chemical pyrolysis of carbon-containing raw materials was developed by Russian specialists.

2. Laboratory and pilot-plant studies of acetylene production natural gas and some types of coal were carried out. This method for treating raw materials was found to be more effective than the traditional mode of production.

3. Pilot-plant tests on a composition of industrial installations of organic production synthesis were carried out using a universal plasma-chemical module.

4. This plasma chemical system was realized in industry.

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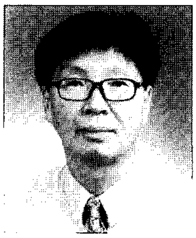

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