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REVIEW

THE STAGES OF RESEARCH ON CREATING COMMERCIAL UNITS FOR PROCESSING OIL SHALE FINES. DEVELOPMENT OF THE "GALOTER" PROCESS IN 1944-1999

E. VOLKOV
G. STELMAKH

Joint-Stock Company G. M. Krzhizhanovski
Power Engineering Institute (JSC *ENIN*)
19 Lenin Av., Moscow
117927 Russia

General Premises, History

The necessity of processing fine fractions of oil shale (oil shale fines in a range of particle size 0-25 mm) constituting up to 70 % of mined shale led to the development of two directions in technology:

- Processing of oil shales in the units with solid heat carrier (SHC) (Russian abbreviation UTT)
- Processing in the units with fluidized bed (FB). It should be noted here that these directions in no case contrast with the widely known and well-developed process "Kiviter", where large lumps of oil shale (25-150 mm) are processed in the retorts with the capacity for oil shale up to 1000 t/day.

The first technological direction mentioned above has been developed and is being improved at the G. M. Krzhizhanovski Power Engineering Institute (*ENIN*), at present the Joint-Stock Company *ENIN*, while the second one has been developed at the Estonian Oil Shale Research Institute (Kohtla-Järve).

At present, two commercial units for processing oil shale fines are in operation in Estonia at Estonian Power Plant in Narva. These units with solid heat carrier SHC-3000 (UTT-3000) have a capacity up to 3000 t/day of oil shale each.

It is too early yet to speak about industrial realization of the FB method for oil shale pyrolysis because the pilot demonstration unit built in Kiviõli (Estonia) at the Oil Shale Chemical Plant (OSCP) *Kiviõli* is temporarily closed down because of the financial reasons in spite of the positive technological results of its operation. Therefore, it is still premature to compare it with SHC by economical, ecological and other characteristics. The same can be said about the method of so-called high-speed oxidative pyrolysis of pulverized oil shale developed at Saratov Polytechnical Institute.

The research and experimental-industrial works carried out for creating the technology "Galoter" are reviewed in the present paper in the chronological order.

The investigations on using a solid heat carrier for semicoking (pyrolysis) of brown coals, peats and oil shales begun during the end of World War II in 1944 in the G. M. Krzhizhanovski Power Engineering Institute of the Acad. of Sci. of the USSR under the leadership of D.Sc. .Eng. I. Galinker. The authors of this process later obtained the patents, and the process when applied to the pyrolysis of oil shales was named "Galoter" (after the family name of one of the authors).

The principle of using hot ash (burnt oil shale) as a solid heat carrier was first described in literature by D.Sc. I. Galinker and a corresponding member of the Acad. of Sci. of the USSR A. Chernyshov [1]. The same article was later published in full in the book by A. Chernyshov [2]. Somewhat later the company *TOSCO* (USA) and thereafter the company *Lurgi* (Germany) patented their technical solutions of this principle. Ceramic balls with $d = 0.5''$ are used in the process "TOSCO-II" as a solid heat carrier, ash or sand - in the Lurgi process. A rotary drum represents the pyrolysis reactor in the process "TOSCO", and it differs from the "Galoter" drum; in the Lurgi-Ruhrgas process the reactor is a combination of two helicoidal mixers and a bunker.

Today these technologies for processing oil shale fines are the most advanced ones and used in industry. Other postwar technical solutions (the process "Gasoter" in the former USSR [3], a process developed in Israel [4], and also "Tasiuk" from Canada-Australia [5]) are still at the stage of pilot and/or demonstration plants.

We have some information about the beginning of the realization of the project "Tasiuk" in Australia for the first pilot unit with the capacity up to 6000 t/day of oil shale fines, its construction is supposed to be ended by the end of 1999.

The process "Petrosix" in Brasil [6] with a large-tonnage capacity (about 2500 t/day for one unit) remains aloof. The lowest limit of processed shale particles is 6 mm, though retorts similar to Estonian retorts "Kiviter" intended for processing lumpy oil shale are used in this process.

Thus, only large-tonnage units SHC-3000 for processing oil shale fines are in use in the world on the commercial scale.

Research and development of SHC technology when applied to oil shales have been carried out for many years in a number of stages. In addition to *ENIN*, the scientific leader, a large number of Estonian organizations and enterprises (Institute of Chemistry of the Estonian Acad. of Sci. (at Tallinn Technical University now), Institute of Economics, Oil Shale Research Institute), and some other institutions of the former USSR have taken part in carrying out the investigations and developments of the projects. Leningrad Department of the State Research and Design Institute *Atomenergoproekt* (*LO TEP*, now Sankt-Petersburg State Research and Design Institute - *SPb AEP*) - general designer of SHC units, chief engineer of the project M. Petrov; Leningrad Branch of the Design Institute *Orgenergostroi*, *Lengipro-neftekhim*, Tallinn Special Design Office and others carried out the design works.

The participation of Estonian specialists was quite natural because before World War II oil shale mining and processing industry was developed on a large scale only in Estonia and Leningrad district. Before 1941, the main object of oil shale processing was the production of liquid fuel, while in the post-war time it was the production of domestic gas for Leningrad and Tallinn. Later on, in the 1960s, oil shale processing was re-oriented to the oil shale chemistry. In the postwar time at the territory of Russia the oil shale processing plant in Slantsy (Leningrad district) was restored. A small factory for producing ichthyol from sulfur-rich oil shales of Kashpir deposit in Volga basin has been working since the 1920s. In the territory of Estonia, the world's largest oil shale processing enterprise in Kohtla-Järve (now *Viru Keemia Grupp* AS [7]) and the largest oil shale chemical plant in Kiviõli worked.

The development of oil shale pyrolysis process and its implementation demanded a long-time research - numerous tests in laboratories and at exprerimental bases, at the pilot and demonstration units for getting the results on processing oil shale samples from the deposits of the former USSR and from other deposits of the world.

Laboratory, Pilot and Experimental-Industrial Investigation of the Processes Taking Place in the SHC Units

In the postwar time, laboratory units representing enlarged retorts with outside heating and supply of solid heat carrier, specially adapted to pyrolysis, and also periodically and continuously working bench-scale and pilot plants which partly or in full reproduce the SHC technology were created in *ENIN* and its experimental bases in Tallinn and Kiviõli (Estonia), at Verkhnesinevid (Ukraine), and Saratov (Russia). Standard laboratory investigations (Fischer retort, calorimetric bomb, etc.) were also carried out. Laboratory units were designed by E. Shapatina, G. Ter-Oganesyan [8], D. Vorona; the retorts by G. Krasnovsky [9], and, in last years, a modernized enlarged retort with the capacity of about 1 dm^3 was designed by K. Yorudas. A. Samoilov developed the express-method of pyrolysis, which enables to study the process using only a few grams of oil shale. Single elements of the SHC technology were studied at the mentioned units, while the study of the full pyrolysis process and equipment used was carried out at the bench-scale units of continuous action.

From 1946 to 1992, the works on creating the process "Galoter" were conducted by B. Tyagunov.

The first pilot unit with a capacity of oil shale 2.5 t/day (~100 kg/h) created with the participation of D. Vorona and E. Grigoryeva was built in 1947 and it worked up to 1956 in Tallinn at the engineering plant *Ilmarine*.

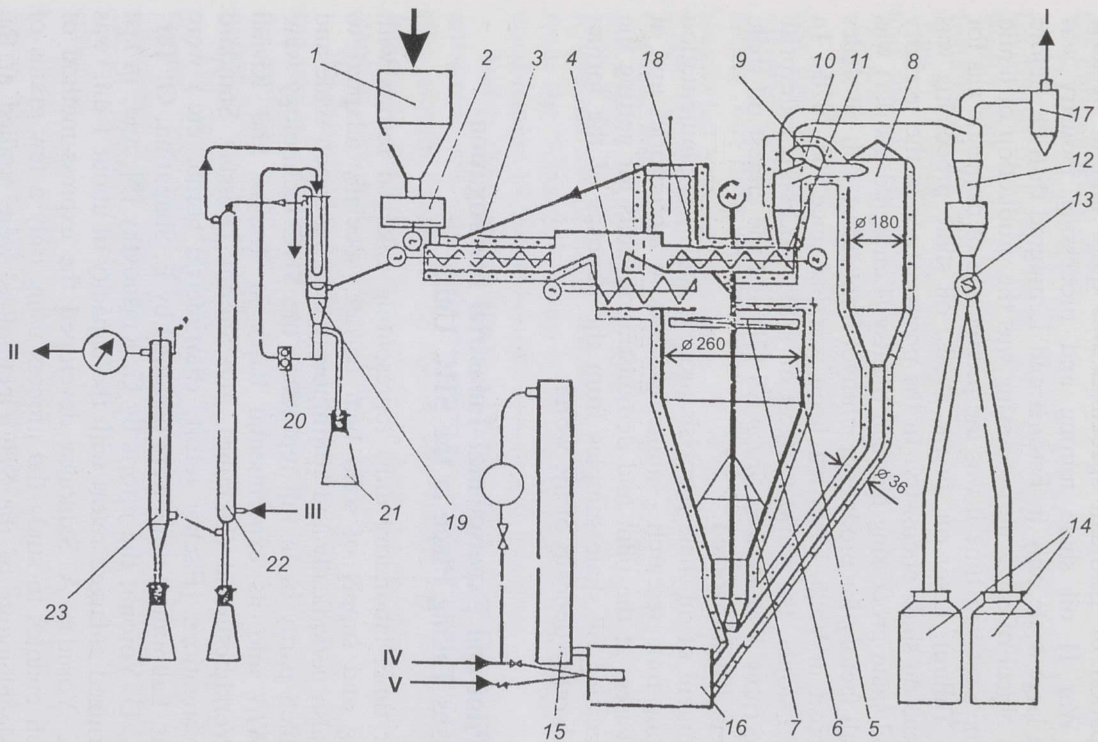

Fig. 1. Bench-scale unit "Kiviõli": *1* – service bunker; *2* - feeder; *3* - dry shale conveyer; *4* - mixer; *5* - reactor; *6* - level controller; *7* - distributing cone; *8* – heat regeneration furnace; *9* - by-pass; *10* - heat carrier separator; *11* - heat carrier conveyer; *12* - 1st stage ash separator; *13* - switching device; *14* - receiving hopper; *15* - air heater; *16* – lighting-up furnace; *17* - 2nd stage ash separator; *18* - fine filter; *19* - scrubber; *20* - sprinkling pump; *21* - receiver; *22* - condenser; *23* – electric precipitator.
I - flue gas; II - retort gas; III - water; IV - air; V – fuel

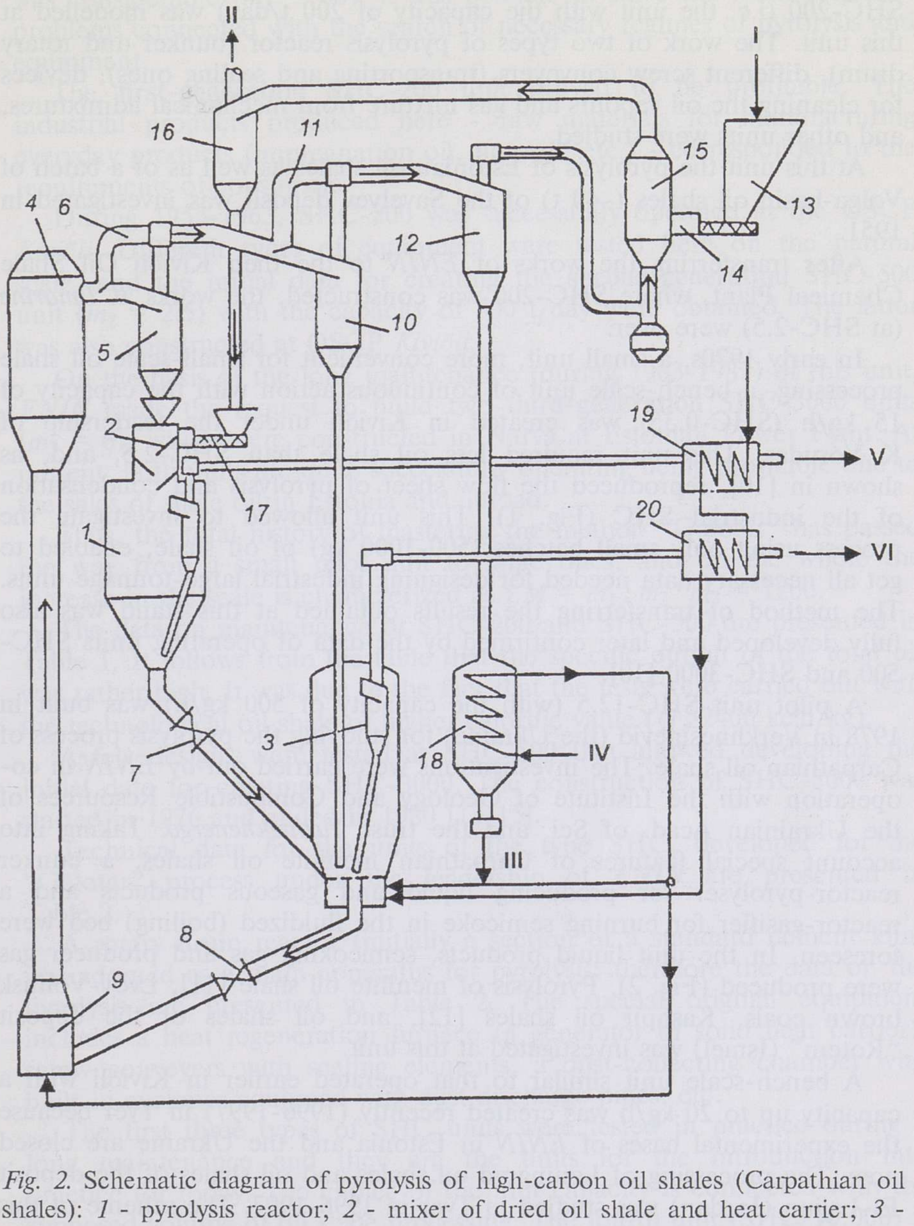

Fig. 2. Schematic diagram of pyrolysis of high-carbon oil shales (Carpathian oil shales): *1* - pyrolysis reactor; *2* - mixer of dried oil shale and heat carrier; *3* - gasifier with fluidized bed and built-in cyclone; *4* — heat regeneration (aerospouting) furnace; *5* — 1st separator of heat carrier; *6* — 1st by-pass gas duct; *7, 8* - control valves; *9* - control gate-feeder for coke-ash residue; *10* — 2nd heat carrier separator; *11* — 2nd by-pass gas duct; *12* - ash separator; *13* — crude shale bunker; *14* - feeding-sealing screw conveyer; *15* - drier; *16* - flue gas cyclone; *17* - sealing screw conveyer of dry oil shale; *18* - ash heat exchanger for air heating; *19, 20* - steam superheaters.

Flows: I - feed shale; II - flue gas; III - ash; IV - air, V —oil vapours and gas mixture; VI - producer gas

Practically all the main equipment of the next demonstration unit SHC-200 (i.e. the unit with the capacity of 200 t/day) was modelled at this unit. The work of two types of pyrolysis reactor (bunker and rotary drum), different screw conveyers (transporting and sealing ones), devices for cleaning the oil vapours and gas mixture from mechanical admixtures, and other units were studied.

At this unit the pyrolysis of Estonian oil shales as well as of a batch of Volga-basin oil shales (~40 t) of the Savelyev deposit was investigated in 1951.

After transferring the works of *ENIN* to the then Kiviõli Oil Shale Chemical Plant, where SHC-200 was constructed, the works at *Ilmarine* (at SHC-2.5) were over.

In early 1970s, a small unit, more convenient for small-scale oil shale processing, a bench-scale unit of continuous action with the capacity of 15 kg/h (SHC-0.35) was created in Kiviõli under the leadership of K. Yorudas. This unit required less oil shale than SHC-2.5, and, as shown in [10], reproduced the flow sheet of pyrolysis and condensation of the industrial SHC (Fig. 1). This unit allowed to investigate the process using only small batches (500-1000 kg) of oil shale, enabled to get all necessary data needed for designing industrial large-tonnage units. The method of transferring the results obtained at this stand was also fully developed and later confirmed by the data of operating units SHC-500 and SHC-3000 [10].

A pilot unit SHC-12.5 (with the capacity of 500 kg/h) was built in 1978 in Verkhnesinevid (the Ukraine) for studying the pyrolysis process of Carpathian oil shale. The investigations were carried out by *ENIN* in co-operation with the Institute of Geology and Combustible Resources of the Ukrainian Acad. of Sci. and the trust *Yuzhtekhenergo*. Taking into account special features of Carpathian menilite oil shales, a bunker reactor-pyrolyser for producing liquid and gaseous products and a reactor-gasifier for burning semicoke in the fluidized (boiling) bed were foreseen. In the unit liquid products, semicoking gas and producer gas were produced (Fig. 2). Pyrolysis of menilite oil shale [11], Lvov-Volinsk brown coals, Kashpir oil shales [12], and oil shales of the deposit "Rotem" (Israel) was investigated at this unit.

A bench-scale unit similar to that operated earlier in Kiviõli with a capacity up to 20 kg/h was created recently (1996-1997) in Tver because the experimental bases of *ENIN* in Estonia and the Ukraine are closed now. The processing of Leningrad oil shales and oil shales of the deposit El-Lajjun (Jordan) was studied in Tver in 1996-1997. The scheme of this unit repeats that of the Kiviõli unit.

Experimental-Commercial Units

As mentioned above, the first-generation experimental-commercial demonstration unit SHC-200 was created at OSCP *Kiviõli* on the basis of recommendations obtained at the unit operated at the engineering plant

Ilmarine in Tallinn. Though the scale transfer from SHC-2.5 to SHC-200 was considerable (m_1 = 80), it was possible to solve the main technical problems connected with the search of necessary regimes of pyrolysis and equipment.

The **first-generation** SHC-200 unit proved to be profitable. The industrial products produced here - raw materials for manufacturing everyday products (impregnation oil, nerosin, etc.) - corresponded to the requirements of consumers.

During 1953-1963, SHC-200 was successfully operated at the OSCP *Kiviõli*. Different types of equipment were tested here on the natural scale, and the initial data for creating the **second-generation** SHC-500 unit (m_2 = 2.5) with the capacity of 500 t/day were obtained. The latter was also constructed at OSCP *Kiviõli*.

On the basis of the long maintenance (during 1963-1981) of this unit, *ENIN* made the request to build two **third-generation** SHC-3000 units (m_3 = 6), which were constructed in Narva at Estonian Power Plant. At present, both units are quite successfully operating being profitable due to the sales of shale oil in Estonia and abroad.

Thus, the total history of mastering the method "Galoter" has passed the way from a small pilot unit to large ones, and on the whole the increase in the scale is characterized as $\Sigma M = m_1 \cdot m_2 \cdot m_3 = 1200$.

The data on maintenance of SHC-200 and SHC-500 are presented in Table 1. It follows from the table that the specific output of the total oil was rather high. It was due to the fact that the tests were carried out with the technological oil shale of higher calorific value ($Q \geq 2400$ kcal/kg).

A few designs were tested at SHC-500 that allowed developing the initial data for creating SHC-3000. The construction of SHC-3000 was started in 1976 and ended in 1980 [13-15].

Technical data for the units of the type SHC, developed for the "Galoter" process under the leadership of *ENIN* are presented in Table 2.

A rotary drum reactor (initially a section of a standard cement kiln) was adopted as a main apparatus for pyrolysis, therefore the data on the pyrolysis are presented in Table 2. The special original equipment includes a heat regeneration furnace (a generator of solid heat carrier), screw conveyers with sealing elements, a dust-collecting chamber with built-in cyclones equipped with dust discharge pipes, etc.

The first three types of SHC units were tested in practice during a long maintenance and they are the units for the introduction into practice for today. The choice of the unit capacity is connected with the supposed volume of oil shale processing. The fourth unit (SHC-10,000) is now only hypothetical, a calculated one.

All standard sizes of reactors are characterized by the same 50 % degree of filling with the solid material and the same pressure (0.2 kgf/cm^2) in the inside space of the drum reactor.

As shown by practice, transfer to the higher capacity has always required a certain time of mastering, because the designs underwent some changes due to the increase in the scale.

Table 1. **Operating Data for the First and Second Generation SHC Units**

Type of unit	Period of maintenance	Number of operating hours	Amount of processed oil shale, thous.t	Amount of shale oil obtained, t	Amount of gas obtained, mln m^3	Yield of total oil, kg/t
SHC-200	1953-1963	17000	102	15800	>5.0	154
SHC-500	1963-1981	107300	2095	286000	109.0	136

Table 2. **Technical Characteristics of SHC Units**

Type of unit	Diameter of drum reactor, in/out, m/mm	Reactor length, m	Residence time of solid material in drum reactor, min	Residence time of oil vapours and gas mixture in drum reactor, s	Rate of motion of solid material in drum reactor, m/s	Throughput rate of oil shale, t/h,	Speed of drum reactor, rotation, rpm
SHC-200	1.64/2000	4.5	20.6	18	0.35	6.34	1.83
SHC-500	2.52/3000	6.0	21.4	15	0.57	20.2	1.18
SHC-3000	4.38/5000	14.0	20.7	21.2	1.1	139.0	0.92
SHC-10000 (theoretical calcul.)	6.37/7000	21.0	18.0	19.8	1.8	417.0	0.92

Table 3. **Maintenance Data on SHC-3000 Operation at the Energotechnological Department of Estonian Power Plant**

Year	Processed		Output			Duration of operation, hour	Notes
	Oil shale, t	Waste, t	Shale oil from oil shale, t	Shale oil from waste, t	Retort gas, thous. m^3		
1995	506582	-	65352	-	18000	5097	
1996	546101	16400	75823	5757	19800	4960	3 months of overhauls
1997	608889	9500	78444	5002	22150	5651	

For example, six types of dust precipitators after reator - centrifugal (disk, ventilated with the rotational speed up to 3500 rpm) of cyclone type, dust-precipitation chambers and others - were tested at SHC-200 with simultaneous improvement of the dust collection from the oil vapours and gas mixture. The best results were obtained using a dust removal chamber with built-in cyclones.

The built-in cyclones were improved in the conditions of SHC-3000: they were equipped with the dust discharge pipes (the 1st stage) and dust ejector (the 2nd stage). It allowed to obtain, in the department of condensation, heavy shale oil with a content of mechanical admixtures ≤ 1.0 % while the further improvement of the process (application of the secondary pyrolysis of heavy shale oil) enabled to get the total shale oil with a content of mechanical impurities ~0.02 %.

Larger sizes of reactor's rotary drum required also changing the design of seals at the inlet and outlet of the reactor.

At present, the main technological and mechanical problems connected with the maintenance of SHC-3000 could be considered to be solved in principle.

The works on creating and commercial mastering of SHC-200, 500, and 3000 units from 1952 to 1992 were headed by an Estonian honoured engineer, state price winner B. Tyagunov. These works were carried out by the collective of *ENIN* research workers: V. Chikul (responsible for the work), A. Smirnov, J. Tuvikene, G. Krasnovsky and others, and also by the specialists of OSCP *Kiviõli* (J. Ulanen, I. Tänav and others), Institute of Chemistry of the Estonian Acad. of Sci. (A. Elenurm, M. Marguste and others), Oil Shale Research Institute (V. Yefimov, S. Doilov, T. Purre and others).

A large contribution at mastering of SHC-3000 was made by: V. Chikul (the head of the introduction group), M. Petrov, S. Vereshchaka, V. Svetlichny and others together with the management of the Estonian Power Plant (K. Senchugov, M. Gudkin), and the collective of its energotechnological department (A. Popov, A. Kaidalov, V. Kindorkin and others).

Organic waste in an amount of 3 wt.% is regularly processed annually together with oil shale since 1996 at the SHC-3000 units.

The possibility of processing crushed tyres, rubber waste, oil sediments from oil tank wagons, oily soils, etc. has been confirmed in practice. Crushed tyres can be processed in an amount up to 10 wt.% (they contain metal cord), while other wastes - in an amount of 25-30 wt.% - without changing operating conditions of pyrolysis.

Processing of different materials together with oil shale increases the efficiency of SHC-3000 operation (the output of gas and total shale oil).

The results of SHC-3000 unit operation for the last three years are presented in Table 3. The common oil shale with the calorific value $Q =$ $= 2000$ kcal/kg is processed at SHC-3000, and that determines the yield of shale oil and gas, which are somewhat lower than these obtained at SHC-500.

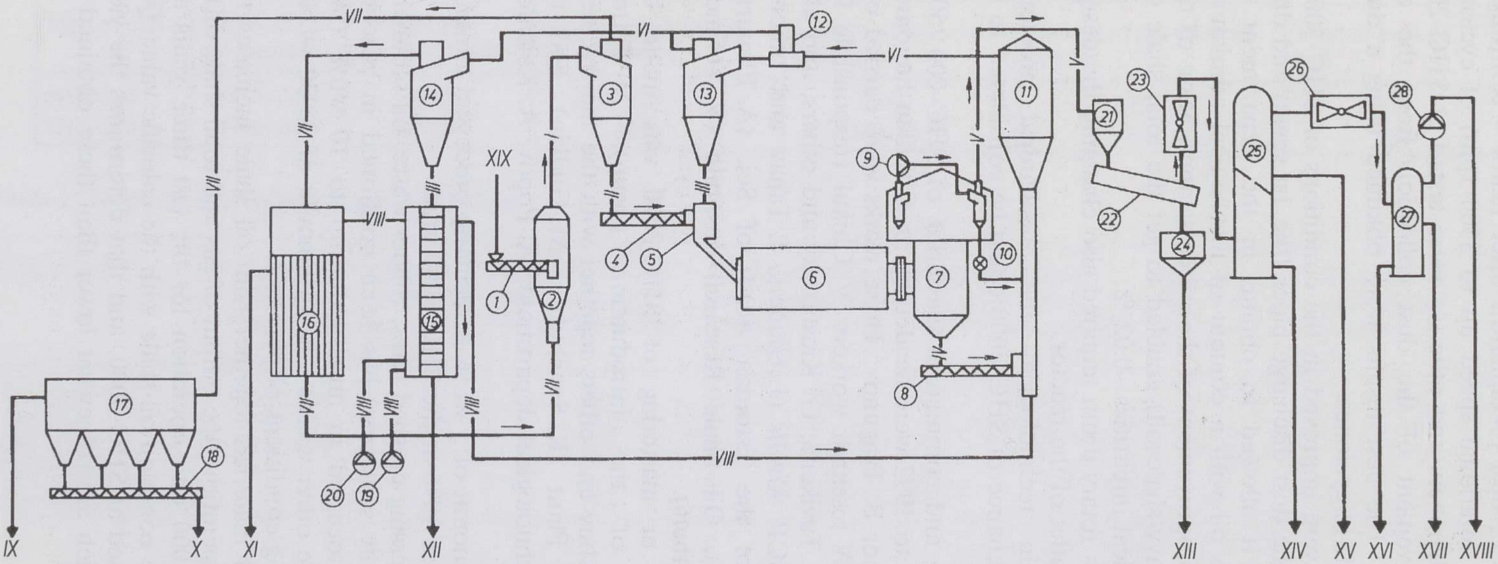

Fig. 3. "Galoter" process, distillation and condensation scheme (schematic diagram of SHC-3000 units): *1* - feed shale conveyer; *2* - drier; *3* - dry shale cyclone; *4* - dry shale conveyer; *5* - mixer; *6* – rotary drum reactor; *7* - dust removal chamber with built-in cyclones; *8* - semicoke conveyer; *9* - fan; *10* - dust recovery system; *11* – heat regeneration furnace; *12* - heat carrier by-pass; *13* - heat carrier cyclone; *14* - ash cyclone; *15* - ash heat exchanger; *16* – waste heat boiler (recovery boiler); *17* - electric precipitator; *18* - dust conveyer; *19, 20* – air blowers; *21* - heavy oil washing tower; *22* - gas collector; *23* - heavy oil cooler-condenser; *24* - heavy oil tank; *25* - rectification tower; *26* - cooler-condenser for gas-naphtha and tar water; *27* - separator; *28* - gas blower.

Flows: I – feed shale mixed with drying agent; II - dry shale; III - heat carrier (ash); IV - semicoke and heat carrier; V – oil vapours and gas; VI - ash in flue gas flow; VII - flue gas; VIII - air; IX – flue gas into chimney stack; X - ash from electric precipitator; XI - steam 40 atm abs, 440 °C; XII - ash; XIII - heavy oil; XIV – heavy oil middle oil mixture; XV - gas-turbine fuel fraction ; XVI - tar water; XVII - naphtha; XVIII - semicoke gas and gas naphtha; XIX – feed oil shale

In addition to Estonian oil shales, Bulgarian oil shales were processed at the experimental-industrial units SHC-200, SHC-500. The oil shales from 20 deposits of the world including Green-River (USA), Stuart (Australia), Timahdit (Marocco), El-Lajjun (Jordan), Rotem (Israel), Alexinac (Yugoslavia), Gurkovo (Bulgaria), as well as shales from Russia, Estonia, Byelorussia, and the Ukraine were tested at the pilot and low-scale (laboratory) units.

The new **fourth-generation** SHC unit is now represented by SHC-3000 designed for the energotechnological complex in Slantsy (Leningrad district, Russian Federation). It has a modernized flow chart and some improvements in the condensation department. The complex will be equipped with four SHC-3000 units.

The main purpose of the fourth-generation SHC-3000 is to obtain the maximum amount of gas turbine fuel to guarantee the most efficient operation of SHC units working as a part of electric power plant in the complex with steam-gas plant (SGP) equipped with gas turbines for both liquid and gaseous fuels.

The flow sheet of oil shale processing including the pyrolysis and condensation departments will have a new solution. A recovery boiler that combines the functions of recovery boiler and ash heat exchanger is suggested for the pyrolysis department instead of two existing cumbersome and metal-intensive units.

The condensation department will work in accordance with the new flow sheet taking into account the fact that the main part of liquid fuel and all retort gas should be used in the gas turbines.

Therefore the whole amount or a part of heavy fraction of shale oil will be directed into the reactor for the secondary pyrolysis together with oil shale. As a result, the quantity of the gas turbine fuel will be increased 2-2.5 times, and all the fraction of shale oil coming from the condensation department will be free from mechanical impurities (<0.02 %).

Technological connections and material flows will undergo corresponding changes, too.

The proposed principal scheme of pyrolysis and condensation departments is presented in Fig. 3.

Among a series of experimental-industrial units created under the leadership of *ENIN*, special attention should be paid to the one (heat regeneration (aerospouting) furnace) erected at Dobrotvorskaya Thermal Power Plant in the of Lvov district (the Ukraine) near the boiler PK-19 with the steam output 120 t/h utilizing Lvov-Volinsk coal.

In this case the question concerns not only the creation of a technology for manufacturing the products from oil shales and coals, but this unit represents a power version of SHC method as the oil vapours-and-gas mixture obtained at pyrolysis of a low-grade fuel together with a solid heat carrier enters the boiler furnace already as a high-calorific qualitative fuel.

The use of such a technology allows power engineering:

- Reduction (3-4 times) of the emissions of dusty fly ash, nitrogen oxides, and sulfur oxides into atmosphere as sulfur will be captured by the alkali components of ash
- Improvement of the action capacities of the boiler
- Use of low-grade fuels whose combustion in boiler furnaces is impossible without adding some highly reactive expensive fuel

This unit was constructed but, regrettably, after 1991 these works transferred to the Acad. of Sci. of the Ukraine were temporarily closed down due to the absence of financing.

A similar version of the heat regeneration furnace was developed and suggested by *ENIN* for the boiler ZKTI-75-39 at Syzran TPP for Kashpir oil shales.

When processing sulfur-rich oil shales or shales containing other undesirable components it is necessary to carry out their pyrolysis together with the subsequent cleaning of obtained products, for example, by means of hydrofining of liquid products from sulfur or cleaning the flue gases after combustion of sulfur-containing liquid fuels from sulfuric anhydride.

In many cases, as the calculations show, the expenses for secondary processes undertaken for modifying pyrolysis products with the aim to protect the environment seem to be economically justified.

Theoretical and Experimental Investigations of Processes Occurring in SHC Units

Theoretical works dealing with special features of the interaction between solid heat carrier and oil shale in the reactor have been carried out since 1944. The collaborators of *ENIN* E. Mirinhoff, O. Tsukhanova, G. Salamandra under the leadership of Prof. I. Galynker formulated the approaches to calculating the warm-up of oil shale by solid heat carrier [17]. Later on V. Solyakov, G. Stelmakh, K. Yorudas, and V. Mamai [18-21] widened the circle of initial premises and approached the development of initial data for a mathematical model to describe the interaction between initial oil shale and solid heat carrier, and determination of the amount of the gas phase allocated during pyrolysis. Theoretical analysis of the processes occurring during mixing solid heat carrier with pyrolized oil shale allowed to calculate the minimum time of warm-up and gas emission, necessary for the establishment of the period of mixture stay in the pyrolysis reactor. The equations of heat and material balance were taken for the basis.

The physical model of warm-up and pyrolysis of particles in the SHC reactor can be presented by a system of equations describing:

- mass exchange processes
- heat exchange processes
- kinetics of chemical processes

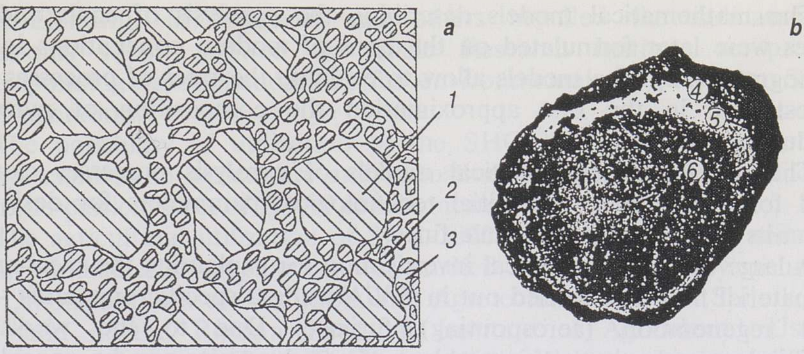

Fig. 4. Bed of the gas-emitting matter in reactor (*a*) and a cross-section of a retorted oil shale particle (*b*): *1* - oil shale; *2* - heat carrier; *3* – oil vapours and gas; *4* - coking layer; *5* – decomposition front; *6* – oil shale core

When studying the heat exchange between oil shale particles and solid heat carrier, it is necessary to pay attention to the following. The phenomenological scheme of pyrolysis typical for SHC and Lurgi reactors is shown in Fig. 4. The size of the predried particles of oil shale is ten times higher than that of the heat carrier particles (500 μm and 50 μm, respectively). When oil shale particles are heated in the reactor of the type "TOSCO II" by ceramic balls, the size of the heat carrier balls will be, *vice versa*, 10-15 times higher than that of the oil shale particles.

In the first case, the heat flow is directed from small particles to large ones, while in the second case - from large particles to small ones. The change in the direction of heat flows depends on the size of heat carrier particles and results in an adequate rebuilding of thermal fields and changes in the nature of fluidization of small particles. In the first case - the small particles are inert, in the other - the small particles emit gas due to the pyrolysis.

In addition, it should be necessary to take into account the fact that the mass of heat carrier always exceeds that of pyrolized oil shale more than two times. It could be not too important for mixing inert materials, however, in thermodestructive reactions heat exchange and fluidization will have different nature.

One may assume that under conditions of mixing large pyrolized oil shale particles with small heat carrier particles the pyrolysis of oil shale particles occurs in accordance with the so-called "front-type" mechanism. The co-ordinates of the particle destruction front in the model (Fig. 4) can be determined, and the velocity of its motion to the centre can be calculated on the basis of the classic solution of the Fourier heat equation for a ball with the fourth-order boundary conditions taking into account the sources and discharges of heat due to the thermal effect of thermodestruction and carrying the heat flow out of the particle by formed oil vapours and gas mixture. Inert particles of heat carrier ash enter the mixing chamber and SHC reactor in the fluidized state, while oil shale vapour-emitting particles enter so the "TOSCO" reactor.

The mathematical models describing the pyrolysis of coals and oil shales were later formulated on the basis of modern conceptions in the monograph [22]. The models allow to evaluate the rates of processes and to estimate in the first approximation the composition of pyrolysis products.

The developed mathematical models of pyrolysis processes can be used for determining the initial technological parameters for designing the units for processing oil shale fines.

A large cycle of theoretical investigation for explaining the mechanism of material flow was carried out in *ENIN* using a specific apparatus - the heat regeneration (aerospouting) furnace. Due to the works of N. Shipkov and others, this problem of gas dynamics can be considered practically solved now [23, 24]. It allows creating the methods for calculating such furnaces [24].

The processes of material flow in a rotary drum reactor were studied by G. Krasnovskii [25].

The calculation methods suggested by G. Krasnovskii for a drum rotary reactor, and by N. Shipkov and A. Perepyolkin for heat regeneration furnace are in fact at present the normative basis for designers of such equipment. These theoretical works were later supported by experimental investigations and tested at the industrial SHC units.

Beside the investigations mentioned above, large attention was paid to the theory of material flow on the transporting-locking screw conveyers. Basing on the experiments, J. Tuvikene developed the methods of calculating such devices [26], fully proved in practice under industrial conditions.

The works on analyzing the flow of dispersed materials in the so-called dust discharge pipes of cyclones in the dust precipitator were carried out in *ENIN* and were very important from the technological point of view. Suffice it to say that the suggested methods of calculation and designing (dust discharge pipes without and with ejectors) allowed to create the units where in the condensation system liquid products with the minimum content of mechanical admixtures can be obtained from the oil vapours and gas mixture. No traditional expensive devices (filters, centrifuges, etc) are needed to purify the products.

A considerable contribution in mastering SHC-3000 was made by the collaborators of *ENIN* A. Gavrilin, Y. Kobsev, S. Vereshchaka, V. Svetlichny, and the collective of the Energotechnological department of the Estonian Power Plant, who developed the theory and introduced into practice the dust discharge pipes mentioned above.

Numerous experimental and theoretical works of physical and chemical character were carried out with the participation of D.Sc. Eng. M. Gubergrits, Ph.Ds A. Kõll, M. Marguste, A. Elenurm, L. Paalme and other Estonian scientists and as well as by V. Mamai and K. Yorudas. These investigations included the study on behavior and disproportionation of oil shale sulfur in the "Galoter" process, the control over forming and binding phenolic compounds - an important problem from the point of view of economics and ecology.

A. Elenurm, M. Marguste and others installed the indicators of emissions, which allowed Oil Shale Research Institute to prepare corresponding recommendations for environmental protection at SHC-3000 operation.

The possibility of utilization of the SHC-3000 ash was studied by many scientists in agriculture and in institutions specialized on building, with the participation of *ENIN* collaborators.

The agricultural utilization of SHC-3000 ash for liming acid soils and as an antibacterial preparation was studied in the Acad. Pryanishnikov All-Union Institute of Fertilizers and Agrotechnics (prof. I. Shilnikov et al.), and in the Russian Research Institute of Agricultural Use of Ameliorated Lands in Tver (T. Filatova with collaborators), as well as in the Estonian Agricultural Academy (under the leadership of E. Turbas), in Lithuanian corresponding institutions, and so on.

The sanitary-hygienic evaluation of agricultural products raised with using the SHC-3000 ash was carried out during several years in experiments on animals in Leningrad Sanitary-Hygienic Medical Institute of the Ministry of Health of Russian Federation under the leadership of Prof. V. Dotsenko by his collaborators [27]. Positive recommendations were finally given.

The investigations on using ash in the building material industry were mainly carried out in the Institute *Giprocement* under the leadership of M. Svatovskaya and L. Fraiman, as well as in the Silicalcite Research and Design Institute (Tallinn) under the leadership of I. Veretevskaya and E. Ojamaa. Positive recommendations on using the SHC-3000 ash were developed in both institutions [28].

Thus, due to the wide front in mastering SHC-3000, the basis for wasteless technology was created. SHC-3000 corresponds to the conditions of ecological safety of such type of anthropological impact.

Economic aspects - efficiency and profitability of the "Galoter" process - have been illustrated by the successful work of two SHC-3000 units of the Energotechnological department at Estonian Power Plant in Narva.

Results of the Bench-Scale Investigations on Pyrolysis of Different Oil Shales

The investigations on processing oil shales from different deposits of the world using the "Galoter" technology were carried out at laboratory installations, in the standard Fischer assay and in enlarged retort.

As a result of the experiments with many oil shales, the "autothermal" parameter K_{eh} (*e* - efficiency, *h* - heat) was suggested by V. Mamai. This parameter expresses the potential heat of the coke-ash residue, which provides the carrying out of pyrolysis at SHC at optimum pyrolysis temperatures without an additional input of heat.

In other words, the pyrolysis of oil shales by heat carrier can be recommended in the cases when the amount of heat remained in semicoke is equal to or exceeds the heat necessary for carrying out thermal decomposition of kerogen without using other heat sources for this purpose. In the case when a considerable amount of heat is remained in semicoke, the remained organics may be subjected to additional gasification or reburning.

The parameter K_{eh} was deduced on the basis of statistical analysis and V. Mamai suggested a nomogram for its practical application (Fig. 5).

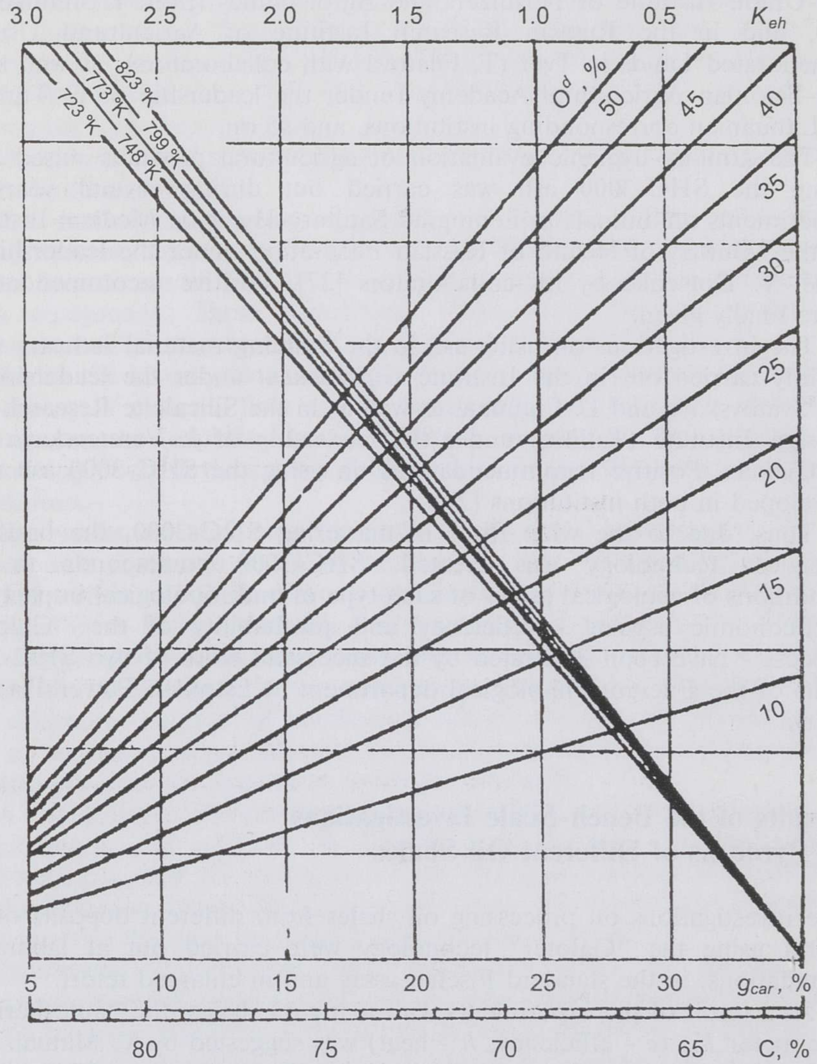

Fig. 5. Nomogram for determination of the possibility for processing oil shales using the method of solid heat carrier: g_{car} - content of coke-ash residue (organic matter basis); O^d - content of organics (dry basis); C - carbon content of oil shale kerogen (dry basis); K_{eh} - parameter of the semicoke "excess" heat

It follows from the nomogram that the pyrolysis process with a solid heat carrier can be realized for oil shales with $K_{eh} \geq 1$, and the process is not suitable without an additional heat supply when $K_{eh} < 1$, i.e. for very poor (low-grade) oil shales with the combustion heat <700 kcal/kg.

According to K_{eh}, all oil shale deposits may be divided into three groups:

- $K_{eh} < 1$ - the heat of semicoke is insufficient for processing oil shales at SHC units
- $K_{eh} = 1.0\text{-}1.5$ - the heat of semicoke is sufficient for the efficient processing of oil shales at SHC units
- $K_{eh} > 1.5$ - in this case there is a heat excess in semicoke which can be extracted through additional gasification (see Fig. 3) or reburning in a special apparatus

The relationship between semicoke quantity per combustible mass and the carbon content C of kerogen has been established analyzing the content and the properties of products obtained in the Fischer retort test for oil shales from more than 30 different deposits. This relationship is approximated by a linear equation:

$$g_{sc} = 121.5 - 1.4C$$

where g_{sc} - semicoke quantity.

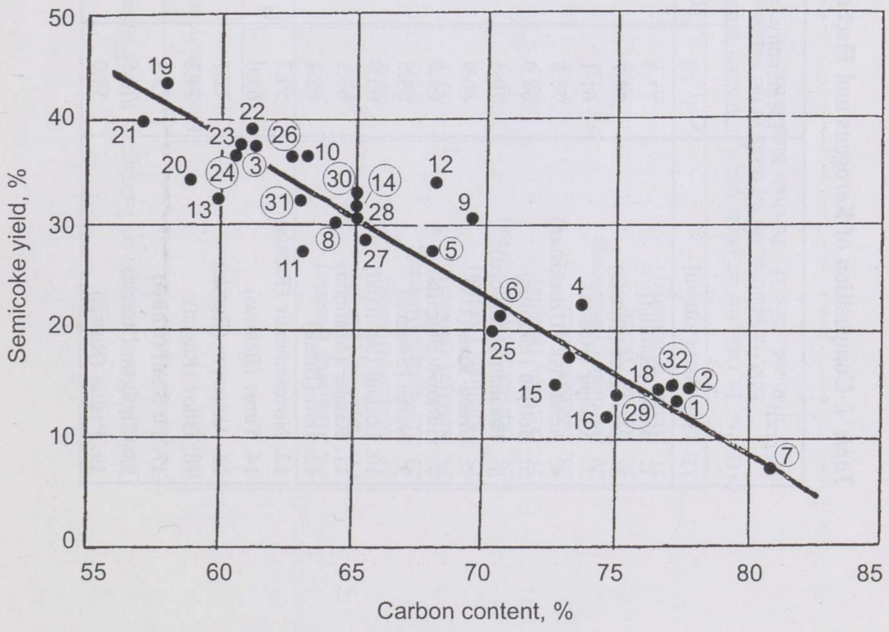

Fig. 6. Semicoke yield versus carbon content of oil shale kerogen; figures at the points denote the number of tests made at different SHC units

Table 4. **Composition of Kerogens and Fischer Assay Product Yields of oil Shales of Different Deposits**

Deposit	Elementary composition of kerogen, %				Fischer assay product yield, % (dry basis)			
	C	H	O	S	Oil	Water	Semicoke	Gas
1. **Baltic** (Estonia)	77.3	9.8	11.2	1.7	65.0	5.4	13.6	16.0
2. **Leningrad** (Russia)	77.7	9.8	11.3	1.2	63.2	5.5	15.2	16.4
3. **Kashpir** (Russia)	61.1	7.3	23.8	7.8	26.6	10.8	37.1	25.5
4. Kenderlyk (Uzbekistan)	73.8	8.4	17.8	-	49.7	11.9	22.8	15.8
5. **Boltysh** (Ukraine)	68.0	9.3	20.8	1.9	43.3	15.3	28.1	13.3
6. **Timahdit** (Morocco)	70.5	9.3	12.4	7.8	40.7	5.2	21.5	32.6
7. **Green-River**(US)	80.9	11.4	6.9	0.8	66.9	8.4	7.9	16.8
8. **Gurkovo**(Bulgaria)	64.3	10.9	24.8		39.2	10.4	31.1	19.3
9. Närke (Sweden)	69.5	7.7	16.8	6.0	28.0	17.0	30.5	24.5
10. Lotiani (Scotland)	63.0	10.1	26.2	0.7	41.0	11.0	37.0	15.0
11. **Rander** (Australia)	63.1	7.9	28.3	0.7	62.2	1.4	27.0	8.7
12. Irati (Brasil)	68.1	10.3	17.9	3.7	40.9	9.8	34.1	15.2
13. Novosemenov (Russia)	59.7	7.8	24.6	7.9	35.8	12.8	31.1	28.3
14. **Turov** (Belorus)	65.0	11.3	23.3	0.4	33.9	14.5	31.0	20.6
15. Alekseyev (Russia)	72.7	8.8	18.5		55.5	14.2	15.1	15.2
16. Gdov (Russia)	74.8	9.0	16.2		63.4	11.8	11.5	12.3
17. Fu Shun (China)	73.4	8.5	10.6	7.5	60.9	4.0	18.0	17.1
18. Chudovo (Russia)	76.6	8.9	14.5		61.0	5.1	14.6	19.3
19. Pashin (Russia)	58.0	9.4	32.1	0.5	24.5	19.8	44.4	11.3

Table 4. Composition of Kerogens and Fischer Assay Product Yields of oil Shales of Different Deposits (end)

Deposit	Elementary composition of kerogen, %				Fischer assay product yield, % (dry basis)			
	C	H	O	S	Oil	Water	Semicoke	Gas
20. **Manturov** (Russia)	58.9	8.3	27.4	5.4	30.1	12.9	34.4	22.0
21. Bogoslov (Russia)	57.2	8.1	34.7		35.9	8.2	40.1	15.8
22. Ulyanov (Russia)	60.8	6.7	27.4	5.1	28.8	14.1	38.8	18.3
23. Levosiyazh (Russia)	60.6	6.8	32.6		29.8	12.4	37.4	20.4
24. **Savelyev** (Russia)	60.5	7.3	32.2		25.4	22.8	36.5	25.4
25. Dergunov (Russia)	70.4	8.4	13.8	7.4	30.1	24.7	20.1	25.1
26. Obshtchiy Syrt (Russia)	62.9	8.1	21.7	7.3	26.7	16.5	37.1	19.7
27. Oznikov (Russia)	65.5	8.3	20.6	5.6	36.6	10.1	28.8	24.5
28. Chernozatonsk	64.3	7.0	21.8	6.9	36.8	12.8	31.1	20.3
29. **Alexinac** (Yugoslavia)	75.0	9.3	15.7		48.1	16.5	14.0	21.4
30. **Rotem** (Israel)	65.0	7.0	15.4	10.7	28.1	20.9	33.1	17.9
31. **Stuart** (Australia)	63.1	9.6	20.4	6.9	32.48	15.53	32.02	19.9
32. **El-Lajjun** (Jordan)	77.08	5.12	17.8		60.77	7.36	15.2	16.6

Notes: In Table the oil shales tested at different solid heat carrier units: laboratory, pilot or experimental-commercial scale, are given in bold. In addition, the oil shales of the following deposits were also tested: No. 33 - **Carpathian** (Ukraine), No. 34 - **Sysol** (Komi-Russia), No. 35 - **Perelyub-Blagodatsk**, No. 36 - **Kamelik-Chagan**, No. 37 - **Kotsebinsk** (Russia).

Table 5. **Yield and Composition of Oil Shale Pyrolysis Products Obtained at Laboratory Units with Solid Heat Carrier [29]**

	Kukersite of Estonian deposit, Narva	Luban deposit, Belarus	Savelyev deposit	Perelyub deposit	Kamelik-Chagan deposit
Product yield (dry basis), %:					
Oil	14.8	7.4	10-12.5	20.6	19.2
Gas naphtha	0.9	0.2	0.1-0.2	0.1	0.3
Gas	5.3	3.1	3.2-5.2	7.9	8.4
Oil characteristics:					
Density, g/cm^3	0.96	0.93	0.97	1.02	1.06
High heat value, MJ/kg	38.4	41.1	-	36.82	38.13
Elementary composition of oil, mass%:					
C	83.9	82.7	79.5	76.3	78.5
H	9.76	10.1	9.3	8.5	8.6
S	0.62	1.3	6.1	8.3	5.9
O + N	5.72	5.9	5.1	6.9	7.0
Cl	0.1	-	-	-	-
Retort gas:					
Density, kg/nm^3	1.165	1.214	0.955	1.346	1.242
High heat value, MJ/nm^3	46.4	37.2	26.8	28.4	27.2
Gas composition, general %:					
CO_2	2.6	18.5	3.9	17.5	26.9
H_2	16.0	21.7	17.5	12.5	23.0
CO	9.6	6.7	10.0	11.6	4.7
C_nH_{2n+2}	31.4	27.7	21.4	15.0	18.8
C_nH_{2n+2}	35.9	24.1	9.4	8.8	9.8
H_2S	0.1	1.3	10.8	34.6	16.8
Nitrogen compounds	4.4	-	-	-	-

The divergence of data in Fig. 6 is connected with special features of mineral and organic components of oil shale and falls in the classified interval of values. This relationship allows using the presented nomogram for the primary evaluation of the possibility of processing a certain oil shale at SHC units.

Experimental results of processing oil shales from 32 different deposits, including 20 investigations made in *ENIN* at different units with solid heat carrier (the names of corresponding deposits are given in bold in the table), are presented in Table 4 and Fig. 6.

It should be noted that though the Fischer test together with the calculations made by using the nomogram give the presentation about the possibility to process a certain oil shale at a SHC unit, it is still necessary to carry out the tests with certain batches of oil shale at a small-scale unit of continuous action when a design solution of the "Galoter" process is needed.

The fact is that the physical and chemical properties of both oil shale and coke residue determine the implementation of a SHC unit. Oil shale fines can be, for example, ground up to the state which leads to depletion of the system by the heat carrier, or fusible characteristics of the mineral residue will be such as to cause slagging, or the bituminization of the predried oil shale could become the obstacle for its motion in the system. When those undesirable phenomena are detected, it is necessary to use additional measures (for example, to add quartz sand), or to develop a special design of the equipment.

The methods developed in *ENIN* for evaluating laboratory results allow to confirm that all known oil shales with the oil outlet ≥70 l/t or $Q \approx$ 900 kcal/kg can be efficiently (with a profit) processed using the SHC technology whereby qualitative gaseous and liquid fuel products are produced.

As for the results of experimental investigations, it is expedient to give Table 5, including the data on the yield and composition of some special products obtained at the bench-scale unit in Kiviõli [29].

As mentioned above, due to closing the laboratory basis of *ENIN* in Estonia, an analogous unit of continuous action with the capacity up to 20 kg/h was recently constructed in Tver (Russia). At this bench-scale unit the investigations of oil shales from the Leningrad deposit, Jordan (the deposit El-Lajjun) and the Kashpir deposit were carried out under the leadership of K. Yorudas. The unit was modernized, however, the main technology and equipment have remained without changes.

The tests with oil shales from the Manturov deposit (Kostroma district) are intended for the present.

The future Development of Oil Shale Processing in Units with Solid Heat Carrier in Russia

The investigations on creating an oil shale processing complex (SPC) in Slantsy (Leningrad district) have recently been carried out basing on the experience of SHC-3000 operation in Narva (Estonian Power Plant).

The Saint-Petersburg organizations (*Atomenergoprojekt* with the participation of *Giproneftekhim* and *Giproshakht*) basing on the proposals made by the collaborators of the Joint-Stock Company G. Krzhizha novsky *ENIN* have worked out two versions of such a complex. In the first stage, three units SHC-3000 and a 300 MW Power Unit for processing 2.5 mln t of oil shale a year are to be erected.

The feasibility study has been accorded with the nature-protection organizations of the Leningrad district. The question of beginning the construction should be solved before the year 2000. Now the problems of both financing and attracting of investors are being dealt with.

The SPC in Syzran produces ichthyol. In this complex some separate chains of the present-day production are foreseen to be reconstructed, in particular, oil shale reactors will be replaced by an solid heat carrier unit of the scale SHC-500 to keep the production and the range of products on the existing level. It is supposed to add the production of the thiophene concentrate. The reconstruction and modernization of the complex will improve the ecological situation in the region and also increase the profitability of the enterprise.

The implementation of combined processing of crushed tyres and other organic wastes in the mixture with oil shales is of a certain interest.

The experiments at the commercial SHC-3000 unit at Estonian Power Plant gave positive results. The addition of organic wastes in an amount of up to 20 wt.% increases the output of shale oil and gas and is also useful from the standpoint of environmental protection in case of processing oiled soils, etc. [31].

It is possible to mention as prospective works of the JSC *ENIN* investigations and technical and economic evaluations on creating the energotechnological complexes utilizing oil shales of Israel and Jordan.

As shown by laboratory experiments and calculations, there is a real possibility of creating complexes including oil shale processing at SHC-3000 and producing of electricity even when high-sulfur oil shales are used.

Within the frameworks of the present review, it is impossible even to mention all the investigators and all the works made for the creation of the process "Galoter".

Suffice it to say that the bibliography on the process "Galoter" - SHC consists of more than 270 publications written by the research workers of *ENIN* (75-80 %), of Estonian research institutes and other participators.

In addition, there are about 30 descriptions and comments in different reference books and editions written by the authors from the former USSR, Russia and foreign countries, not directly connected with the development of the process "Galoter". During the period when this technology was created, about 40 authorized certificates of the USSR, and 15 foreign patents in 7 countries (1977-1998) were received.

Twelve USSR authorized certificates were re-registered as patents of the Russian Federation. At the present time some new patents are in the stage of formation, and some patents are already given [32, 33].

We regret to say that the majority of people who have been taking part in the creation of the process are not with us now: B. Tyagunov,

M. Gubergrits, Yu. Aranovich, V. Chykul, A. Smirnov, Y. Shelestin, A. Kõll, G. Krasnovskii and other Russian and Estonian scientists.

Some people have deviated from oil shale matter. Many designers, project chief engineers and other participants have changed their field of activity. The Oil Shale Processing Complex in Kiviõli who contributed greatly to mastering SHC has also been re-oriented after the year 1981.

The authors apologize for not mentioning all of the participants in this review. The directorial office of the JSC *ENIN* thanks all of them for their hard labour during many years on investigating and mastering the process "Galoter". Our special thanks belong to the member of the Estonian Acad. of Sci. I. Öpik, who has paid much attention to the mastering of the process "Galoter", supported its creation and pointed out shortcomings which were eliminated as much as possible.

This review is devoted to the memory of **B. Tyagunov** (1910-1992) — an unforgettable enthusiast of the "Galoter" technology, the research leader and one of the main authors of the SHC units.

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