From resinous wood distillation to hydrogen energy (questions of the history of pyrogenation)

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Abstract: Substantial contribution to development of electrical engineering was made in the 19th century. Indeed, in the early 19th century centralized gas lighting systems were established first in England, and then in the rest of Europe and North America, and then it was switched to electric lighting by 1880s. Before electricity became widespread and suitable for general public use, gas was the most popular method of indoor and outdoor lighting in cities. The article is devoted to the history of development of thermochemical technologies for the decomposition of wood and other carbon-containing materials (coal, oil, shale, peat) in order to obtain heat and related valuable products for various purposes. Special attention is paid to the development of gasification technologies of low-quality solid fuels, featuring periods of increasing demand for similar technologies and periods of a sharp decline. As a result of the analysis of world experience in the field of gasification technologies, it is shown that, generator gas theoretically can be used for energy purposes, for example to drive automobiles or internal combustion engines, to be the fuel of CHP plants of different capacity, as well as to be included into the schemes with fuel cells for the development of hydrogen energy.

1. Pioneers in gasification
Resinous wood distillation is probably one of the most ancient mass technologies with perhaps only alcohol distillation that could compete with it. The experimental development of thermochemical processes associated with the decomposition of wood and other carbon-containing materials (coal, oil, shale, peat) began in ancient times, long before the concept of pyrogenation united them. In addition to generating heat associated processes of obtaining a number of valuable technological, medical and even cosmetic products were mastered. Coke made the achievements of the Bronze Age possible and, likewise, tar boosted the development of navigation.

Detailed review of the development of thermal gasification from antiquity to the present days made by V. Kopytov [1] highlights that the number of individual users of combustible gas for lighting is rapidly growing starting from the Middle Ages. In the review it is anticipated that gas was emitted during thermal decomposition and specifically during dry distillation of fuel.

Industrial production of gas for lighting and heating began in the first decade of the 19th century. Britain claimed to lead in this area, as in 1800-1802 William Murdoch on the basis of his own scientific research of the technology of coal gasification provided gas lighting of the Boulton & Watt company, where he was employed. At the same time French engineer Philippe LeBon studying distillation of wood and coal understood that gas produced in distillation could be useful for lighting, heating, and as the energy source in engines. LeBon designed a distillation oven known as a thermolamp that was used...
widely for years. Practically at the same time, specific proposals on the organization of industrial gas production for fuel needs were made in Belgium, a little later in the USA, Canada, and Australia.

In Russia the task of thermal decomposition of solid fuel for lighting was investigated by Pyotr Sobolevsky. Together with a retired lieutenant D’Orer they invented their own thermolamp. In 1812, Sobolevsky and D’Orer, according to the decree of Alexander I, were awarded the Order of St. Vladimir of the 4th degree “for the care and work that they invested in designing the thermolamp which had never existed in Russia before” [1]. In 1813 Sobolevsky inspired by the ideas of LeBon made the gas lighting of the Admiralty Prospect in St. Petersburg. Later this project lost financial support as it didn’t show economic benefits. Sobolevsky gradually lost interest in gasification, focused on metallurgy and became one of the founders of powder metallurgy, and in 1830 was elected the member of the St.Petersburg Academy of Sciences for his achievements.

2. Development of gasification technology

2.1. The first gasifiers in the world

The first gasifiers appeared in 1840s. As independent devices of continuous action, they were used to service various industries and technological processes. The most famous constructions of that time were gas generators designed by German engineer Bischof (1839), French engineer Ebelman (1840), and Swedish scientist Ekman (1845). But gas generators’ application in industry started only after F. Siemens introduced the regenerator principle of heating plant furnace (1861). This principle made it possible to use generator gas efficiently. Indeed, fuel consumption of plant furnace decreased by several times. Thus, brothers F. and W. Siemens invented the first industrial gas generator.

Characteristic design features of Siemens gas generators were the following: the movement of oxygen carrier and the forming gases proceeds from the bottom to top; stepped inclined grate, generation of the required amount of steam by evaporation of water injected under the grid. This design turned out to be so successful that it was widely used for 40-50 years. More sophisticated designs appeared only at the beginning of the 20th century.

Starting from the 1840s practically everyone who was seriously engaged in the research of gas generator considered the possibility of obtaining with its help not only lighting or heating, but also useful technical work. Their efforts were initially doomed to failure, since they were based on the erroneous idea of Huygens about the possibility of obtaining useful work due to atmospheric air pressure. After D. Watt showed the need to increase the pressure of the working fluid and began to produce efficient steam engines, the inventors of gas engines received a new impetus, again mistakenly believing that now all issues were resolved. Moreover, for a long period of time it remained unclear that the work of increasing the gas pressure is several orders of magnitude higher than when generating steam of the same pressure, and that the gas engine has only external similarities with steam engine (cylinder and valves), but on the whole they are completely different engines that implement, as we say now, different thermodynamic cycles. Among the failures on that rather long journey, the most famous were: Lebano (1804), de Rivas (1806), Wright (1833), Barnet (1838, g.), Drake (1842), Barzanti and Mattuchchi (1854-1860) [1]. The last one among them was J. Lenoir, who in 1961 patented an engine, sometimes called three-stroke, because it didn’t have the compression stroke. Being initially doomed to failure, Lenoir widely advertised his “new, lightweight, compact multi-purpose engine” and organized its industrial production. He managed to sell about 500 of these engines before consumers realized that they were “gas-eaters” with an efficiency of about 1 %. In that time the efficiency of steam engines had already reached 15 %, so no one needed such engines. Trying to avoid commercial collapse, the unlucky businessman invited a talented self-taught mechanic Nicholas Otto, instructing him to “modify” the design. Let us give Otto’s qualifications its dues as by his efforts the efficiency of the Lenoir engine exceeded 3 %. Further, Otto was luckier as he empirically discovered the important role of the compression stage for the gas engine and understood the four-stroke cycle of the heat engine. N. Otto was the first one to build a workable four-stroke engine. He patented his discovery and in 1864 launched successful production of engines, with products of combustion of gas obtained from coal as the working fluid. In terms of efficiency, Otto engines were not inferior to steam engines, but they were much lighter, simpler, more reliable, and there was a steady demand for them [2]. Continuing to improve his engine,
Otto increases its efficiency to 22 % by 1878, which was higher than the efficiency ever achieved by steam engines. It is not surprising that in the following decades such engines became widespread. In this regard, it is interesting to note that in 1860 Siemens brothers proposed a gas engine, involving a gas generator unit with a four-cylinder piston engine, and even began its testing [3], so the idea can be confidently said was in the air.

Electric power industry evolved in the middle of 1880s and grew rapidly. Gas engines were in strong demand to drive electric generators (which were then called "dynamo"). By the end of the 19th century unit power of gas engines reached 1000–1200 kW. However, the use of gas derived from plant biomass for its drive was steadily declining because preference was given to cheaper fuels such as coal and oil refining. The share of plant raw materials in the balance of energy fuel gradually reached only 2 %, although interest in burning peat and shale remained.

Initial period of power systems development is characterized by the absence of any serious environmental restrictions; smoking chimneys were perceived as a sign of technical progress and expectations of a soon-to-be universal well-being.

The first visible result of electric power industry development was the rapid reduction of gas lighting, that disappeared by the middle of the twentieth century. Gas plants with powerful gas holders were no longer needed. Gas generators were saved only where it was a part of the technological process (metallurgy, ceramic production, oil and slate industry, etc.). In spite of this the development of designs and methods for the operation of gas generators continues. New technical solutions for grates, loading systems and slag removal appeared, various types of blowing systems, forms of gas generator shaft, methods of dealing with fuel arching and much more were tested [4].

2.2. Russian experience

In the beginning of 20th century use of generator gas for households was studied at St. Petersburg Polytechnic Institute. This work was supervised by professor Vladimir Karlovich Valgis. He studied the gasification of peat and shale on an experimental gas generator (gasifier). As a result, by 1920 he proposed the technology of shale gasification and the idea of gasification of Petrograd on its basis. But the main reserves of shale remained in Estonia which became independent by that time. The first Estonian gasifier was launched in Kohtla-Järve in 1924. After 1945 the plant in Kohtla-Järve was restored, its capacity increased, which made it possible from September 1948 to supply Leningrad with domestic gas produced from Estonian shale. Only in 1959 natural gas replaced the generator gas. Apparently the era of using generator gas in everyday life was over. As for V. K. Valgis, we only know that in the early 1930s he led the work on hydrogenation. This process allowed obtaining liquid petroleum products from brown and black coal. Such technologies were then successfully developed in Germany and the United States.

2.3. Transport gasifiers

For a while, interest in gas engines operating on wood remained in transport. This was a period when automobile engines were extremely imperfect and it was not clear whether automobile industry would take the path of steam, gas, gasoline or electric engines. As soon as gasoline engine succeeded in this motor competition, interest in gas-generating engines weakened for a while, so that it would soon be revived, but for entirely different reasons.

The wide spread of liquid fuels in transport gave rise to certain counter process. Countries deprived of petroleum resources saw an alternative to liquid fuels for transport in gas derived from various organic materials, primarily wastes. In forest abundant regions it also seemed more profitable to use local raw materials instead of imported ones. In 1920, German-French engineer G. Imbert developed transport gasifier, where gas and fuel moved in the same direction. Such a gasifier was called the downdraft (Imbert) gasifier [5, 6]. During operation with such a scheme of movement, the incoming air burns and pyrolyzes some of the wood, cracks most of the tars and oils, and burns some of the charcoal that fills the gasifier below the nozzles level. This allowed increasing the gas calorific value, simultaneously clearing it of tar, which favorably affected the engine. As a result, since the 1920s works on the creation of gas-generating engines received a new impetus. A number of quite successful technical solutions appeared, and by the end of the 1930s a lot of gas-generating vehicles were in operation in many
countries. For example, in the USSR by 1945 10 % of all cars were gas-generating, while the share of gas-generating cars and tractors in the forest industry was close to 100 %. As an example, a soviet car is shown in Fig. 1. At the same time gas-generating fleet (mainly river boats and tugboats) and prototypes of gas-generating locomotives and even tanks were created. Only by the mid-50s, as a result of the development of oil refining and pipeline transport, the program of gas-generating cars was curtailed, and in 1962, the production of stationary gas-diesel engines stopped.

Figure 1. A gar GAZ-42 (GAZ-42) (produced in the USSR from 1938 until 1949).

2.4. Synthetic fuels
By the 1920s appeared a number of technologies essential for the development of bioenergy. In 1926, German researchers Franz Fischer and Hans Tropsch discovered a chemical reaction in which in the presence of a catalyst (substances containing iron or cobalt) a mixture of gases CO and H₂ was converted into various liquid hydrocarbons. This mixture got the name of synthesis gas. The discovery was important because it indicated the path to the production of synthetic fuels. The first industrial reactor using this technology was launched in Germany in 1935. Then production of coal from synthetic gasoline, synthetic diesel fuel, and lubricants based on the Fisher-Tropsch technology was launched in Germany, Japan, China, the USA, South Africa, etc. Until the beginning of the 1990s, the FT technology plant operated in Novocherkassk (Russia), where various chemically pure products were obtained from the coal of Donetsk Basin and later from natural gas. FT technology has come a long way of modernization and improvement; the name “Fischer-Tropsch” is now applied to a large number of similar processes: Fischer-Tropsch synthesis, Fischer-Tropsch chemistry, etc. [1].

In the middle of 1920s, F. Winkler developed the first gasifier with the “boiling” layer, which opened up a new promising direction in the organization of furnace and drying processes. Already in the early 1930s, the German company Lurgi designed the first fixed bed gasifier operating under a pressure of 3 MPa, which made it possible to intensify significantly the gasification process, at the same time reducing the size of the gasifier. This method of gasification was called the Lurgi process [1, 2, 6, 7].

A number of studies conducted in the USSR in the 1930s–1960s contributed to the expansion of the current level of knowledge. Efficient designs of gasifiers with a direct and indirect process were created at the All-Union Thermal Engineering Institute (VTI) and the Central Research and Design Institute for Mechanization and Energy of the Forest Industry (TSNIIME). V.S. Naumov created a number of progressive automotive-tractor gasifiers; V. A. Lyamin investigated the effect of pressure on the gasification processes of plant biomass and developed his own design of a stationary gasifier at Leningrad Forestry Academy (LTA). In subsequent years, the development of research at LTA’s scientific school was the study of high-speed (flash) pyrolysis [8] and systematic studies of lignin pyrolysis conducted at VNII hydrolysis [9].

The notion that not only solid fuel of plant origin, but also coal, would soon give way to cheap oil and gas, and Fisher—Tropsch’s (FT) synthesis and pyrolysis processes would henceforth be of interest only for wood chemical production was widely spread at that period of time.

3. Global Energy crisis of 1972-1973 and its influence on energy sector
The situation radically changed as the global energy crisis swept across the planet in 1972-1973. Oil prices almost tripled (from about $12 to $42 per barrel), and the world community faced the problem of
oil replacement for the first time. Then the term “renewable energy sources” appeared and the interest in biomass as renewable energy source revived. At the same time, unexpectedly, the knowledge and experience gained by previous generations in the field of handling solid fuels and, in particular, plant biomass turned out to be in demand. However, plant biomass had a special role in the list of renewable energy sources. At that level of civilization development the main part of oil produced (about 60%) was used for the organic synthesis of many essential substances and products. But the same products could be synthesized from a mixture of gases (CO + 2H₂) obtained from coal, using well-known and well-developed technologies.

Then, the majority of developed countries, primarily USA, Canada, Germany, and Great Britain allocated considerable funds into large-scale ambitious research projects related to the development of synthesis gas production technology from solid fuels. Almost all energy giants (Royal Dutch Shell plc, Ahlström Oyj, Lurgi Energie und Umwelt, Chevron Texaco, etc.) and small firms, some of which were specially founded for the purpose, joined to find the solution to the problem. [10].

After a few years the energy crisis was over and the urgent need oil replacement was gone, but by that time the scale of problems caused by environmental pollution became more evident. Therefore, already launched projects, that had obtained first scientific and practical results, continued their work changing focus to the solution of global environmental issues, namely the reduction of the impact of energy on the environment.

It resulted in the creation of powerful gasifiers for coal gasification and the construction of large power plants with the capacity of 250–600 MW in the USA, the Netherlands, Spain, and Italy with gasification of low-grade coal. The most powerful station with the capacity of 950 MW appeared in South Korea in 2000. At the same time technologies for centralized production of liquid and gaseous motor fuels from solid fuels were successfully developing. In many projects, energy production was combined with obtaining valuable chemical products and energy production [11].

This time a new stimulus for the development of technologies and techniques for the energy use of plant biomass and various organic wastes appeared. In the early 1980s the Finnish company Bioneer Oy and the VTT Research Institute conducted large-scale studies on the gasification of wood chips, logging and granulated household waste, as well as peat and straw on a pilot Bioneer gasifier with a thermal capacity of 1.5 MW. These investigations enabled the launch of mass production of similar gasifiers of 3–5 MW thermal capacity. The gasifiers have been successfully exploited in Finland and Sweden to produce heat. Subsequent modifications of Bioneer gasifiers are still produced by Finnish firms Condens Oy, Carbona Oy and a number of other firms. [12].

4. Modern gasification technologies and gasifier application

At the end of the 1980s an integrated gasification combined cycle (IGCC) power plant was proposed and received wide commercial support for municipal solid waste (MSW) treatment. The main scientific and technical problems that determine the feasibility of IGCC implementation for waste were associated with generator gas cleaning, which governs the efficiency and normal operation of a gas turbine wheel space. Currently, these problems are mostly solved. Programs for the implementation of IGCC were conducted in the USA (IGT), Finland (Tampella, VTT), Sweden (TPS). Gasifiers differ in design, however, almost all of them implement circulating fluidized bed technology, which, unlike fixed bed gasifiers (similar to Bioneer), is characterized by complex aerodynamics of two-phase and two-component flows. To control such a process, an advanced system of regulation and automation is required. In addition, for successful operation of these gasifiers, special fuel preparation (sorting, granulation, etc.) is necessary and qualified service personnel is required [13]. According to the design studies, IGCC cycle in the range of electrical power from 50 to 150 MW allows to obtain an efficiency of 45–50% and a coefficient of fuel utilization of 90%. At the same time, emissions into the atmosphere are 20–50% lower than with traditional combustion methods.

In Russia state program for the development of renewable energy sources appeared only at the beginning of the 1990s, including a project on the energy use of biomass was announced [14]. Research group of St. Petersburg State Technical University together with "Energy Technology" and CNIDi research institutes designed and tested a number of gasifiers with a capacity of 50, 600 and 3000 kW working with various types of plant biomass and waste (Fig. 2). This scientific collaboration resulted in
studied modes of joint operation of gasifiers and diesel engines, developed schemes of thermal power plants operating on plant biomass and waste [11]. The Institute of Problems of Electrophysics and Electric Power Engineering of the Russian Academy of Sciences conducted studies on plasma pyrolysis of highly toxic waste [7].

![Figure 2.](image)

**Figure 2.** Fixed bed gasifiers designed and tested: a) G-3 gasifier (capacity 3 MW), b) G-50 gasifier (capacity 50 kW), c) UTG-600 gasifier (capacity 600 kW).

In subsequent years, the number of organizations and researchers involved in has increased. The bulk of work was devoted to fixed bed gasification and the improvement of fixed bed gasifiers. This work was carried out at the Institute of Thermal Physics (ITF) of the Siberian Branch of the Russian Academy of Sciences and at the Ural Federal University [15]; at Kazan Technical University [16] and [17]; in the MMPP Salyut [1] and the Institute of Problems of Chemical Physics (IPC) of the Russian Academy of Sciences (in Chernogolovka), and etc. Several fundamental publications are devoted to this topic [11].

Ideas of distributed energy and production of standard fuels from waste are developed at the Institute of High Temperatures of the Russian Academy of Sciences [18], specifically issues of obtaining hydrogen and pure carbonaceous materials based on gasification [19]. Original studies of the production of liquid fuels based on wood hydrogenation are carried out at LTA and Arkhangelsk University of Technology [8].

Research into the energy use of plant biomass is also being actively pursued at Lappeenranta University of Technology (LUT), Finland, under the supervision of Professor Esa Vakkilainen. Currently LUT scientists have focused on improving the properties of biomass through technologies such as torrefaction [20] and hydrothermal gasification [21]. Both processes occur at relatively low temperatures. The resulting biochar (bio-torrefaction and hydro-coal, respectively) have a higher carbon content and calorific value than the original biomass. Significant difference between the processes is that when torrefaction of the reaction occurs in the atmosphere of inert gas at a pressure close to atmospheric, whilst in hydrothermal gasification, the raw material is in aqueous suspension under pressure, usually in a saturated state.

The experimental facilities of the LUT allowed scientists to understand the mechanisms of the processes and to develop models suitable for further research. The main objective of these studies is to understand the impact of the initial biomass quality on torrefaction and hydrothermal carbonization, to outline the possibility of integrating these technologies into thermal power plants, as well as to figure out the main factors affecting the effectiveness of this integration.

Recent decades have determined the main areas of research related to the energy use of plant. Sometimes they are quite distant from each other other both in scientific base and in technical methods of implementation; however, they are united by the fuel and raw material base and the desire to quickly solve the energy problem of humanity [22, 23]. Among them are small, medium and high capacity power units (up to 150 MW) [23], generating electrical and thermal energy from waste and mixed fuels based on direct or two-stage combustion and gasification technologies; improvement and optimization of
equipment (furnaces, gasifiers, fuel handling systems and gas cleaning); design and optimization of
distributed power plants like CHP with low capacity and diesel power plants [11, 21].

5. Might gasification and hybrid technologies be combined?
The problem of creating hybrid energy complexes, combining the installation of hydrogen energy and
the installation of gasification technology is being studied in detail in the world. The implementation of
the relevant processes requires the presence of free hydrogen H₂, the production of which is associated
with high costs of electrical energy [24].

One of the most promising ways for the development of hydrogen energy is based on the use of fuel
cells, which allow direct conversion of the energy of a chemical reaction into electrical energy. The
reaction occurs with the continuous supply of fuel (hydrogen) and oxidant (air). Such fuel cells are called
electrochemical generators (EG).

It can become the basis for providing distributed consumers with electrical energy and also can be
considered as an element of promising large power plants. In the latter case, we are talking about hybrid
plants, where due to the optimal combination of traditional thermodynamic cycles with electrochemical
generators, a further increase in the efficiency of the production of electrical and thermal energy is
ensured. At the moment the cost of these electrochemical generators is high, primarily due to the use of
expensive materials (platinum, etc.). The possibility of reducing the cost of EG is primarily associated
with high-temperature solid oxide fuel cells operating at temperatures of 850–1000° C, as well as with
molten carbonate fuel cells, whose operating temperature is in the range 650–850° C.

The advantages of solid oxide fuel cells are that they do not require expensive catalysts and can run
on different types of fuel. The limiting efficiency of EG operating on oxygen-ionic electrolytes can reach
60 %, and with the use of proton electrolytes one can expect to obtain an efficiency of about 80 %. Recently, a number of studies have been devoted to the thermodynamic analysis of hybrid schemes [25],
however, coal is usually considered as the source fuel. Improvement of technologies for the production
of composite fuels based on biomass, organic waste and local fuels makes the research of specific
features of hybrid installations based on EG relevant for the tasks of distributed energy and for
improving high-power units.

It can be expected that in the very near future the problem will be solved, and then the power industry
will take another qualitative leap resulting in the significant change in power equipment structure. The
framework of an operating single power complex of a hybrid plant will include stages of gasification of
plant biomass, hydrogen separation and purification, the generation of electrochemical electricity, the
generation of heat necessary for the implementation of electrochemical processes and the development
on the basis of its utilization of additional energy.

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