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HISTORICAL REVIEWS

HISTORICAL OVERVIEW OF USING FLUIDIZED-BED TECHNOLOGY FOR OIL SHALE COMBUSTION IN ESTONIA

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The use of oil shale as a fuel in Estonian Power Plants started at the beginning of the 20th century (e.g. Tallinn Power Plant 1923, Püssi Power Plant 1937). For combusting lump oil shale grate-firing technology was used. The limitations of grate-firing technology forced to search for or to create better combustion technologies. In these years the pulverized-firing combustion technology was already successfully used for coal combustion. Estonia was the first country to implement the oil shale pulverized-firing combustion technology in the new Kohtla-Järve Power Plant (in operation since January 1949). Almost at the same time in the Institute of Industrial Problems (IIP) of the Academy of Sciences of the Estonian SSR (later the Institute of Termophysics and Elektrophysics (ITEF)) and then the Estonian Energy Research Institute (EERI) the experimental and theoretical research of oil shale combustion in fluidized bed was started. In 1948 Dr. Sc. Hans Truu, who was the vice-director of IIP and the head of the Department of Furnaces (later the Department of Combustion Processes (DCP)) introduced the subject of fluidized-bed combustion technology in Estonia. During more than fifty years the fluidized-bed combustion of several oil shales (from Estonia and Syria, Morocco and Volga basin), other fuels (Estonian oil shale semicoke, dictyonema argillite, wood, peat, coal) and their mixtures (Estonian oil shale with semicoke, oil shale with dictyonema argillite, oil shale with coal, wood with peat) was studied along with other technologies (e.g. pulverized-firing of oil shale) up until the incorporation of EERI into Tallinn University of Technology in 2004. During this fifty years eleven scientific theses were defended (seven of them dealing with fluidized-bed combustion technology) and several projects implemented by the scientists of the DCP of the EERI. This article gives a short overview of using fluidized-bed technology for oil shale combustion, studied in IIP (ITEF and EERI).

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Introduction

The first fluidized-bed combustor with heat output of 0.15 MW_{th} was completed in the Department of Furnaces of IIR in 1949–1950 under the guidance of Hans Truu, and the first oil shale combustion tests were carried out in 1951 [1]. The first results of oil shale combustion tests in this fluidized-bed combustor were successful and formed the main part of his doctoral thesis. To curry out industrial tests of this technology the reconstruction project of a 10-MW_{th} boiler with mechanical grate in the Püssi Power Plant was worked out. In 1957, the year H. Truu died, the reconstruction of the boiler was completed. In 1959 the boiler was fired up, and the oil shale combustion tests lasted up until 1962 [2, 3]. After industrial combustion tests the laboratory tests of Estonian oil shale and other fuels in fluidized bed continued in TEFI [4, 5]. A new laboratory oil shale fluidized bed gasifier with heat output of 0.2 MW_{th} was constructed, basing on the experience of the industrial and laboratory experiments [6].The construction of the new oil shale gasifier was protected by the Author's Certificate USSR 709938 [7].

For industrial tests for oil shale combustion with gasification in fluidized bed a new fluidized bed gasifier with heat output of 35 MW_{th} was worked out [8]. For unknown reasons the implementation of the project in the Kohtla-Järve Power Plant was not completed.

The first laboratory fluidized bed combustor

The layout of the first experimental fluidized-bed furnace for burning oil shale fines successfully tested in 1951 is presented in Fig. 1.

The main dimensions of the experimental fluidized bed furnace were: diameter of the gasification chamber 205 mm and height – 980 mm, grate area – 0.0288 m^2 and the part of free flow of the grate area – 1.3%; diameter of the burning chamber of gasification products – 340 mm and height – 1000 mm.

The process parameters of the experiments of the first laboratory fluidized bed furnace are given in Table 1.

The process parameters of the experimental fluidized-bed furnace (fuel load, primary and secondary air amount) were varied on a large scale. It enabled to determine temperature limits (margins) of oil shale bubbling bed. The lowest bed temperature was higher than the oil shale inflammation temperature, and the highest temperature was lower than the fuel ash sintering temperature. In the fluidized bed coarse coke and a small amount of gas were burned to get heat for oil shale gasification. In the temperature range of bubbling bed 568–1035 °C the apparent heat load of the grate was 2.28–4.61 MW/m^2. The gasification products and fine coke particles were burned in the burning chamber with secondary air.

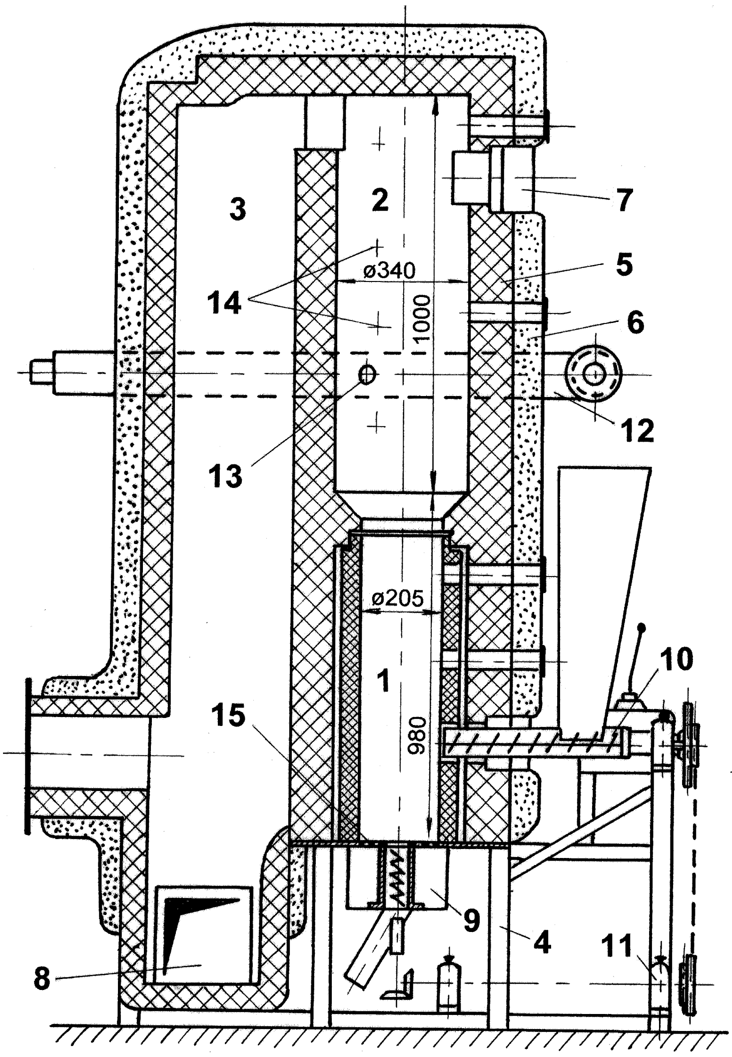

Fig. 1. The first experimental fluidized-bed furnace.

1 – oil shale gasification chamber, 2 – burning chamber of gasification products and fine coke particles, 3 – chamber for separation of fly ash, 4 – framework, 5 – chamotte refractory, 6 – asbestos wool, 7 – explosion flap, 8 – hatch, 9 - primary air collector with grate and ash removal screw, 10 – fuel feeder, 11 – gearing of fuel feeder and ash removal screw, 12 – secondary air pipe, 13 – nozzles of secondary air, 14 – holes for measurements of process parameters and for extraction of gas and ash samples, 15 – grate.

The main results of the first experiments of oil shale combustion in the laboratory fluidized bed furnace were as follows:

- The usability of the fluidized-bed technology for oil shale combustion was successfully proved.

Table 1. **The process parameters of laboratory experiments**

Parameter	Unit	Result
Oil shale calorific value as received, Q^r_i	MJ/kg	8.22–16.6
Oil shale max particle size, d_p	mm	3.5
Fuel load, B	kg/h	22.5–44.4
Excess-air rate in oil shale gasification chamber, α_g	–	0.39–1.09
Overall excess air rate, α	–	1.06–1.46
Thickness of fluidized bed, L	mm	400–450
Bed temperature, t_b	ºC	568–1035
Temperature in the burning chamber of gasification products and fine coke particles, t_f	ºC	872–1166
Apparent heat load of grate, q_g	MW/m^2	2.28–4.61
Calorific value of gasification products, Q_g	MJ/nm^3	2.8–5.7
Medium velocity of gases in oil shale gasification chamber, w_g	m/s	0.77–2.80
Bottom ash, A_b	%	28–60

- High content of the volatile matter of oil shale (average content in used oil shale was 82.5%) enabled to use the gasification process of oil shale in fluidized bed. It allowed significantly to decrease the grate area.
- For oil shale a new combustion technology was proposed. Oil shale is gasified in the fluidized bed, and the gasification products and fine coke particles are burned in a separate furnace.

The first industrial fluidized-bed combustor

Good results of laboratory tests encouraged to carry out industrial tests of oil shale combustion with gasification in fluidized bed. Under the guidance of Dr. Sc. H. Truu a project for reconstruction of a 10-MW_{th} boiler with mechanical grate in the Püssi Power Plant was worked out. In the process of reconstruction the mechanical grate of the boiler was removed, and the fluidized bed gasifier and the burning chamber of gasification products and fine fly coke were inserted in front of the boiler. In 1957 the reconstruction of the boiler was completed.

The cross-section of the fluidized bed gasifier and the combustion chamber of gasification products and fine fly coke of the reconstructed boiler in the Püssi Power Plant is presented in Fig. 2.

The grate of the fluidized-bed gasifier was made from cast iron plate. The grate area was 2.8 m^2 (width 1 m, length 2.8 m) and the open-grate area made 2.3% of it. The air box under the grate was divided into three parts along the grate length; it enabled to regulate the primary air input into different parts of the fluidized bed. The inclination angle of the grate was 5º in the direction of the end. Gasification products and fine coke particles were burned in the combustion chamber with secondary air. For better separation of ash from gases the combustion chamber was designed in the form of horizontal (semi)cyclone. The combustion products were cooled in the furnace and in the convection pass of the boiler, purified from fly ash in the cyclone and directed into the atmosphere with the help of the flue-gas exhauster.

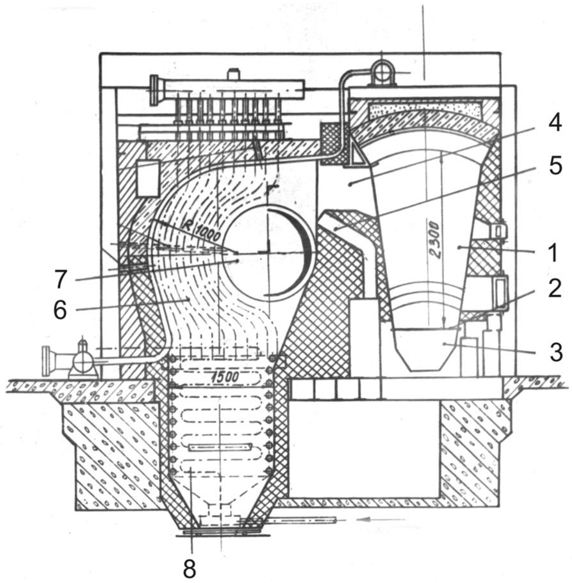

Fig. 2. Cross-section of the reconstructed boiler [2, 3].

1 – fluidized bed gasifier, 2 – grate, 3 – air box of primary air, 4 – exit hole of gasification products and fine fly coke, 5 – secondary air channel, 6 – combustion chamber of gasification products and fine fly coke, 7 – exit hole of combustion products into boiler furnace, 8 – combustion chamber hopper.

Experimental

In 1958 the first team from the Department of Furnaces Endel Jürismaa (head), Ants Martins and Roland Karjus started the fieldwork in the Püssi Power Plant to fire up the reconstructed boiler. It took about one and a half year to amend and improve some components of the reconstructed boiler. In 1959 the boiler was fired up and oil shale combustion tests lasted up until 1962 [2].

Oil shale used for experiments (max particle size 3–5 mm) was screened out from oil shale used in the Püssi Power Plant furnaces. It took about a week to fulfil the hopper of the reconstructed boiler with screened oil shale (about 90 t) sufficient for one experiment with duration maximally 30 hours. If there had been a stationary oil shale preparation system, the reconstructed boiler would have worked continuously.

For preheating of the fluidized-bed gasifier wood (about 30–40 kg) was burned on the grate, which was covered with oil shale ash. After burning of wood oil shale was led into the gasifier. The general start-up time of the boiler lasted 1.5–2.5 hours, after that the steam from the boiler was directed into the steam turbine.

The oil shale was led into the gasifier by screw conveyor at the input to the grate. By the general idea of that time the fuel, canalised into fluidized bed, had to mix very quickly with whole bed material. Actually the mixing of fresh oil shale particles with bed material was of local character at the input to the grate. The bed material (fuel, coke and ash particles) moved on along the grate like a fluid, with speed determined by fuel load. Due to the horizontal movement of the bed material, oil shale gasification took place in the first zone of the bed, burning of coarse coke particles occurred in the second zone, and the third zone worked as the hot ash cooler (or air heater). The medium thickness of the fluidized bed was regulated by the height of the grate barrier.

The preliminary tests showed that the area 2.8 m^2 of the fluidized-bed grate was designed too large for the boiler in operation, as coke was completely burned out in the second zone at the full load of the boiler. Therefore the last part of the air box was closed and the grate area decreased to 1.8 m^2. The process parameters of the experiments of the industrial fluidized-bed furnace are given in Table 2.

Table 2. **The process parameters of the experiments of the industrial fluidized-bed furnace**

Parameter	Unit	Result
Oil shale		
– moisture, W^r	%	12.7–16.7
– ash, A^d	%	43.8–49.5
– carbonates, $(CO_2)^d_M$	%	13.1–15.7
– calorific value, Q^r_i	MJ/kg	9.6–10.6
– max particle size, d_p	mm	3–5
Fuel load, B	t/h	2.75–4.72
Excess-air rate in oil shale gasification zone, α_1 (primary air)	–	0.35–0.45
Excess-air rate in gas and fine fly coke burning chamber, α_2 (secondary air)	–	1.2–1.7
Thickness of fluidized bed, L	mm	300–400
Bed temperature, t_b	ºC	840–875
Temperature in the burning chamber of gasification products and fly coke, t_g	ºC	1350–1050
Apparent heat load of grate, q_g	MW/m^2	4.7–6.8
Calorific value of gasification products, Q_g	MJ/nm^3	4.2–6.7
Medium velocity of gases in oil shale gasification chamber, w_g	m/s	1.35–1.53
Actual thermal output of the boiler, D	MW	6.0–9.4
Ash balance		
– ash from fluidized bed gasifier, A_g	%	23–25
– ash from burning chamber of gas and fine fly coke, A_{bc}	%	62–63
– ash from furnace and from convection part, A_{cp}	%	4.5
– ash from cyclone, A_c	%	1.0–.5
– ash loss (into the atmosphere), A_a	%	6.0–9.5

There was no need to use heat exchange surfaces in the bed. In the gasification zone temperature level of the bed was very easy to regulate with the amount of primary air (α_1 = 0.35–0.45). In the coke combustion zone the temperature was higher (895–958 °C) than that in the gasification zone, but much lower than the sintering temperature of the bed ash (about 1050 °C).

The combustion process of gases and fine fly coke was very intensive in the burning chamber due to high temperature of gases and very good mixing of gases and secondary air. Therefore sometimes the temperatures in burning chamber exceeded the temperatures of ash sintering and melting. To prevent this process the amount of cold secondary air was increased (α_2 = 1.4–1.7) and frequent ash separation from the burning chamber was provided.

Aerodynamic separation of coke and ashes particles took place in the fluidized bed gasifier. The content of CaO + MgO in the bottom ash from the gasifier was 45–50% and that in the bottom ash from the combustion chamber 36–39%. The decomposition rate of carbonate compounds of the oil shale mineral part in ashes from the fluidized bed was 82–97% and in bottom ashes from the burning chamber 85–98%.

Basing on the first industrial tests of oil shale combustion with gasification in the fluidized bed the following observations attracted attention:

- The lengthwise motion of the bubbling fluidized bed along the grate demonstrated the obvious formation of the gasification zone of oil shale and the combustion zone of coarse coke particles.
- The rate of ash separation from the fluidized-bed gasifier together with the ash from the burning chamber formed 85–88% of fuel ash.
- The combustion process of the gases and fine fly coke in the burning chamber was very intensive due to high temperature of gases and very good mixing of gases with secondary air, and therefore sometimes the temperatures in the burning chamber exceeded the temperatures of ash sintering and melting.
- Depending on aerodynamic separation, the chemical content of the mineral part of ashes from the gasifier and from the combustion chamber differed substantially. It brought about the fact that the melting temperature of the bottom ash from the burning chamber was lower than that of the ash from the fluidized bed.

The new bench-scale oil shale fluidized-bed gasifier

After completing oil shale industrial combustion tests in the Püssi Power Plant, the laboratory combustion tests of Estonian oil shale and other fuels continued in EEI.

A new laboratory oil shale fluidized bed gasifier with heat output of 0.2 MW_{th} was constructed, basing on the experience of industrial and laboratory experiments [4–6]. The diagram of the new gasifier is given in Fig. 3.

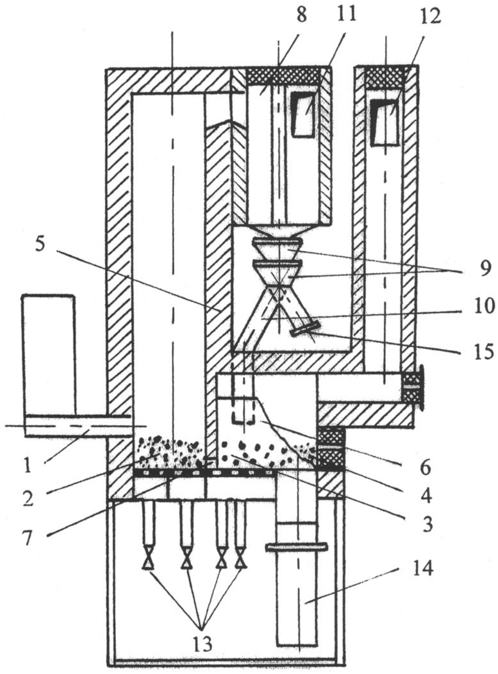

Fig. 3. The laboratory oil shale fluidized bed gasifier with heat output of 0.2 MW_{th}.

1 – fuel feeder with hopper, 2 – oil shale gasification zone, 3 – coarse coke combustion zone, 4 – fine coke combustion zone, 5 – dividing wall between oil shale gasification zone and coke combustion zones, 6 – barrier between coarse and fine coke combustion zone, 7 – hole for coarse coke transfer from the gasification zone into the combustion zone, 8 – fly coke separator, 9 – coke valve boxes, 10 – tract of fine coke, 11 – exit window of cleaned hot gases, 12 – exit window of coke combustion products, 13 – primary air, 14 – ash hopper, 15 – damper for ash samples.

The main modernizations of the new test bench were:

- dividing wall 5 between the gasification zone 2 and coke combustion zones 3 and 4;
- transfer of coarse coke into the combustion zone through the hole 7 in the dividing wall 5;
- cleaning of hot gases in the fly coke separator 8;
- canalisation of separated fly coke back into the fluidized bed in the coke combustion zone;
- combustion of coarse and fly coke in different combustion zones in 3 and 4, separated by the barrier 6.

The air for combustion of coarse and fine coke was introduced into fluidized beds from different air boxes. It enabled to regulate the appropriate air speed for fluidization of coarse and fine coke and to prevent the secondary elutriation of fly coke and fly coke ash from the fluidized bed.

The construction of the new oil shale gasifier was protected by the Author's Certificate USSR 709938 [7].

The principle objectives of the tests were:

- to assess suitable temperature level for oil shale gasification in the fluidized bed and sulphur capture in the fluidized bed and in the freeboard on the one hand;
- to guarantee complete combustion of coke residue in the fluidized bed and to avoid the ash sintering process in the gasifier on the other hand [9, 10].

The chemical analysis of the used oil shale was the following: Q^d_i = 11.75 MJ/kg, $(CO_2)^d_k$ = 18.93%; S^d = 1.7%; A^d = 48.40 %; particle size – 0–3 mm.

In the oil shale gasification zone the fluidized bed temperature was regulated only by the amount of primary air. In the range of the excess air rate from 0.28 to 0.53 the temperature in the fluidized bed rose from 800 to 950 °C. At the same time the calorific value of hot gases decreased from 7000 to 4000 kJ/m^3. The temperature in the gasification zone was regulated only by the amount of primary air. The dependence of calorific value and temperature of hot gases on the excess air rate is presented in Fig. 4.

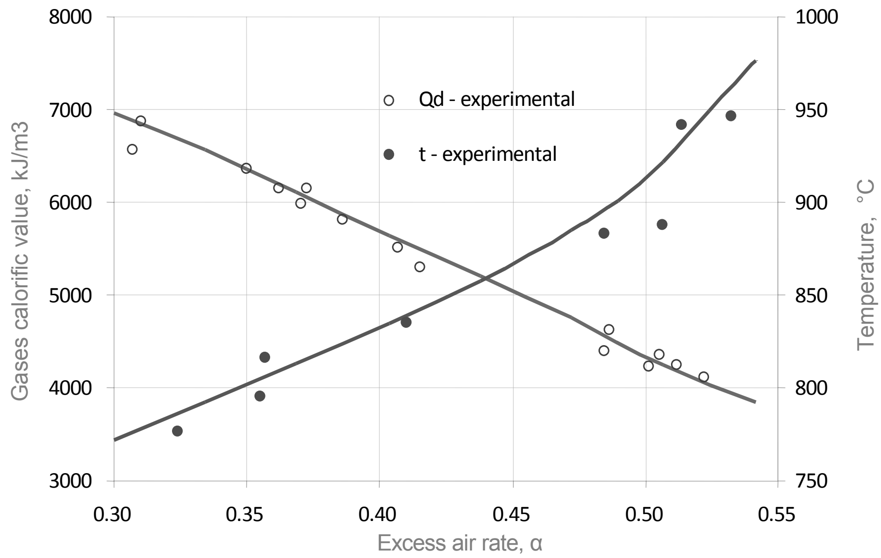

Fig. 4. Dependence of calorific value Q of hot gases and temperature on excess air rate α.

The apparent heat load of the grate in the gasification zone was 3.9–5.9 MW/m^2. The apparent heat load of the fluidized bed gasifier grate was controlled by Formula 1 [11]:

$$Q/F = q_a{\cdot}w_0/\alpha,\ \mathrm{MW/m^2}, \tag{1}$$

where

Q – heat supplied into the fluidized bed with fuel, MW;
F – fluidized bed grate area, m^2;
q_a = Q^r_i/W_0, MJ/m^3 (Q^r_i – calorific value of oil shale as received, MJ/kg,
W_0 – theoretical amount of dry air for complete combustion of oil shale, m^3/kg);
w_0 – air speed, m/s;
α – excess air rate.

The bed temperature is one of the most sensitive parameters for sulphur capture in the fluidized-bed combustor. The other parameters are the presence of sorbent (Ca, Mg) and time for chemical reaction. The mineral part of oil shale contains a sufficient amount of carbonate compounds to bind the sulphur in it, in oil shale the molar ratio Ca/S = 7–8. The ratio Ca/S = 2.5–3 is sufficient for the total capture of sulphur [12].

During oil shale gasification the sulphur in gases was mainly in the form of H_2S. The sulphur capture process in the fluidized-bed gasifier occurred according to the following reactions [13, 14]:
calcination of carbonate compounds of oil shale mineral part in the gasification zone

$$CaCO_3 \rightarrow CaO + CO_2\ ; \tag{2}$$

capture reactions in the gasification zone

$$CaO + H_2S \rightarrow CaS + H_2O \tag{3}$$

and

$$CaCO_3 + H_2S \rightarrow CaS + H_2O + CO_2; \quad .(4)$$

oxidation reaction in the zones of coarse and fine coke combustion

$$CaS + 2O_2 \rightarrow CaSO_4. \tag{5}$$

Maximum sulphur capture rate 0.9 was measured at temperatures from 828 to 850 °C in the fluidized bed and in the freeboard of the gasifier (Fig. 5). During performed tests two main sulphur capture requirements were guaranteed: suitable temperature interval and needed calcium to sulphur molar ratio (Ca/S). However, the time for capture reactions was too short (about 2 s) because of small dimensions of the gasification chamber to achieve the maximum feasible sulphur capture rate. Theoretically it was approved that sulphur capture rate 0.99 could be achieved in case the retention time is 4–5 s [14].

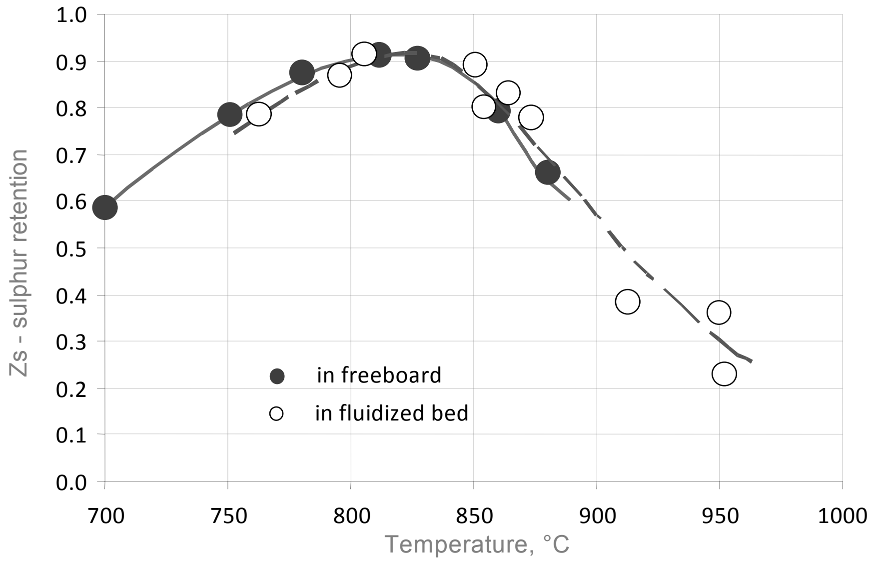

Fig. 5. Influence of temperature on the sulphur capture rate Z_S: 1 – in the fluidized bed and 2 – in the freeboard [14].

The complete combustion of coke and oxidation of sulphides were guaranteed at medium temperature of the fluidized bed 895 °C and excess air rate 1.15–1.25 in the coke combustion zone (Fig. 3–4, 5). 85–89% of oil shale ash was discharged from the gasifier.

Temperature rise in the fluidized bed, especially in the gasification zone in the deoxidation medium, caused ash sintering and collapse of fluidization. Temperatures of ash sintering and softening depend on its chemical composition [15, 16]. Chemical composition of ash fractions is heterogeneous, most changeable is the content of SiO_2 and CaO. Due to aerodynamic separation in fly ash the content of SiO_2 is increased and CaO content is decreased. CaO content of the coarse bottom ash was 41–45% and of the fine bottom ash 33–36%.

A special method was elaborated for determination of the sintering temperature of oil shale ash [17]. This method enabled to determine experimentally the dependence of oil shale ash sintering temperature T_s on the ratio $SiO_2/CaO = m_0$. The increase in CaO content of oil shale ash increases the sintering temperature of ash.

The sintering temperature of oil shale ash is described by empirical Formula:

$$T_s = 1530 - 366 \cdot m_0, \text{ K.} \tag{6}$$

Member of Estonian Academy of Sciences I. Öpik defined the deformation temperature of oil shale ashes by Formula [15]:

$$T_1 = 1730 - 233 \cdot m_0, \text{ K.} \quad (7)$$

From Formulas (6) and (7) it follows:

$$T_s = 1.57 \cdot T_1 - 1188, \text{ K.} \quad (8)$$

Formula (8) could be used for evaluation of sintering temperatures T_s of oil shale ash in the case the deformation temperatures T_1 of these ash samples are measured by the standard method.

The main results of testing the improved model of the oil shale fluidized-bed gasifier:

- Oil shale gasification tests in the fluidized bed enabled to determine the influence of the excess air rate on the temperature in the fluidized bed and on calorific value of hot gases.
- The combustion of coarse and fine coke in separate zones of the gasifier enabled to discharge 85–89% of oil shale ash from the gasifier.
- For the first time it was experimentally proved that the use of the fluidized-bed technology enables to bind oil shale sulphur with carbonate compounds of oil shale ash. The optimum sulphur capture process took place within the temperature range 800–850 °C in the fluidized bed and in the freeboard. The moderate sulphur capture rate 0.9 was achieved because dimensions of the gasification chamber of laboratory test bench were small. Theoretically it was approved that sulphur capture rate 0.99 could be achieved in case the retention time is 4–5 s [14].
- The sintering temperatures of oil shale ash are substantially lower than their deformation temperatures (Formula 7). The temperature rise over T_s in the bubbling fluidized bed caused the collapse of fluidization especially during the fire-up time, when the temperature regime of the bed was not stable.

Combustion of cleaned gases

Hot gases from the gasifier were burned in the compact vertical cyclone furnace [18]. The temperature of gases was 740–870 °C and ash content 43–58 g/nm^3. The walls of the cyclone furnace were cooled by secondary air to get hot air for combustion of gases. The air tubes were covered with refractory bricks. The temperature of secondary air was in the range 555–640 °C. The combustion process of hot gases with hot secondary air was very intensive and the temperature achieved the level of 1420–1640 °C in the furnace. The melted fly ash (low fusion-temperature, $m_0 \sim 1.2$) separated from gases onto the walls of the furnace formed a liquid slag layer and left the furnace in the liquid form.

The main results of the tests obtained from cleaned gases combustion:

- Combustion of cleaned gases in the wet-bottom vertical cyclone furnace guaranteed complete combustion of gases and high separation of ash from combustion products – 95–98%.
- High flame temperature in the furnace caused dissociation of sulphur-ash compounds and along with this the increase in sulphur emission.
- Combustion tests of oil shale gases from the fluidized-bed gasifier confirmed the requirement to burn these gases at lower temperatures. In order to reduce combustion intensity of the hot gas it is necessary to feed secondary air into the furnace in parts and from different heights of the furnace or to use the diffusion-flame gas burners.

Project of 35 MW_{th} oil shale fluidized-bed gasifier

In the Institute of Termophysics and Elektrophysics the fluidized-bed gasifier with heat capacity of 35 MW_{th} for industrial tests of two-stage oil shale combustion method was worked out in 1981 [8]. The technical solution of the gasifier project was based on the ideas presented in the Author's Certificate USSR 709938 [7]. The fluidized-bed gasifier was designed to be used for reconstruction of the boiler No. 3 at the Kohtla-Järve Power Plant.

The oil shale handling system was developed by the researchers of the Chair of Thermal Technology of the Tallinn Polytechnical Institute (now the Institute of Thermal Technology of Tallinn University of Technology) [19].

The technical project of the fluidized-bed gasifier and the reconstruction of the boiler No. 3 were done in the Leningrad Department of *Orgenergostroi* [20]. Eesti Energia (Estonian Energy) financed the project. The fluidized-bed gasifier was planned to be installed ahead of the boiler No. 3 instead of the one-hammer mill in the ash basement. Cleaned gases were planned to be burned in the furnace of the boiler No. 3. For gas combustion the *ENIN*-type diffusion-flame burners of gas were adjusted to guarantee low flame temperature in the furnace. For unknown reasons the implementation of the project was not completed.

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