Oil S67le, 2004, Vol. 21, No. 2
pp. 161-172

ISSN 0208-189X
© 2004 Estonian Academy Publishers

https://doi.org/10.3176/oil.2004.2.07

LABORATORY TEST RIG FOR COMBUSTING ESTONIAN OIL SHALE IN CIRCULATING FLUIDIZED BED

V. DUSHENKO*, H. MERE, I. SHCHEGLOV

Laboratory of Multiphase Media Physics,
Tallinn University of Technology
23A Akadeemia Rd., Tallinn 12618, Estonia

T. TIIKMA**, A. VRAGER

Thermal Engineering Department,
Tallinn University of Technology
116 Kopli St., Tallinn 11712, Estonia

The designed laboratory test rig allows investigating oil shale combustion in circulating fluidized bed. The behavior of oil shale ash micron-size particles was studied with the purpose to estimate the fouling of heat-exchange surfaces. The samples taken from special points of the test rig were analyzed to trace sulphur capture in circulating fluidized bed. There are few successful studies made in full-scale high-temperature conditions on the test rig with circulating fluidized bed. The test data enable, to a certain extent, to prognosticate operating conditions of heat-exchange surface and sulphur capture. The success in understanding oil shale behavior, fouling, and sulphur capture needs future research on combustion in circulating fluidized bed.

Introduction

The key problems in using Estonian oil shale in power plants are: intensive fouling of heating surfaces due to high mineral matter content of oil shale, and emissions of SO_2 and huge amounts of CO_2 liberating at destruction of carbonates at high temperature. These problems can partially be solved by using the technology of circulating fluidized-bed (CFB) combustion [1, 2].

One of the main factors causing the fouling of heat-exchange surfaces is particle deposition onto them. For successful operating of a power equipment it is necessary to know how much solid particles will be deposited from an aerosol flow onto the acting faces. In spite of numerous

---

* Corresponding author: e-mail *vera@eeri.ee*
** E-mail *tiikma@sti.ttu.ee*

investigations of such processes made up to now, there are no satisfactorily reliable methods of estimation of the amount of deposited particles. According to various investigations, there are different reasons of particle deposition onto the surfaces. In general, for developing effective methods of qualitative and quantitative estimation of this process, it is important to know not only the total amount of deposited particles, but also their distribution along the surface, their size, chemical properties, etc.

The investigations of the channel dust-laden flows [3–5] have shown that the Stokes number has a critical value ($S_{t_{cr}}$) defining critical conditions of particle deposition in the wide group of symmetrical dust-laden flows. While such flow meets an obstacle, the particle deposition onto it stops when the Stokes number is

$$S_t = \frac{\Delta^2 \rho_p w}{18 \rho_g \nu d} \leq S_{t_{cr}} \tag{1}$$

where Δ is particle size;

w is gas flow velocity;

ν is kinematic viscosity of gas;

d is cross-flow tube diameter;

ρ_p and ρ_g are density of ash particles and gas, respectively.

Therefore, in this case the coefficient of particle capturing equals zero that is related to the characteristics of the flow at the critical point and defined by the value of the dimensionless gradient of the normal velocity at this point (adhesion condition for the viscous air flow intends zero velocity at the surface). It means that for the given case there is a minimum size $\Delta_{p\,min}$ of particles deposited from the flow. $S_{t_{cr}}$ allows establishing $\Delta_{p\,min}$ of particles which precipitate onto the surface of the given shape – $S_{t_{cr}}$ is greater for other, worse conditions for particle deposition onto this obstacle under the impact of inertial forces. For the given $S_{t_{cr}}$ the particle deposition is more at higher flow velocity and particle density and smaller at linear dimensions of the obstacle.

The researches of Estonian Energy Research Institute at TTU (EERI; since 2004 – Laboratory of Multiphase Media Physics at Tallinn University of Technology) have studied the influence of the parameters of the vertical downward air flow loaded with solid particles streamlining various shapes (flat plate, conical and curved surfaces) on the nature of particle deposition onto the given surfaces. The experiment technique and measuring procedure as well as the data processing technique are described in detail in [6].

Here it must be pointed out that these experiments were carried out under isothermal cold conditions, i.e. in absence of thermophoresis. In actual conditions where the temperature differs in dust-laden flow of gas and on the surface, thermophoresis and adhesion will play an important role in deposition of micron-size particles. Also molecular diffusion induced by the concentration gradient still takes place, and the deposition of particles can occur at $S_t < S_{t_{cr}}$.

Thermal Engineering Department (TED) at Tallinn University of Technology has a rich data bank for solving actual scientific and technological problems related to the use of oil shale in power plants [7]. The data on deposition of very fine oil shale ash particles from high-temperature gas flow given in [8] are contradictory, and additional investigations are of great interest.

Understanding of sulphur capture at fluidized-bed combustion can give an opportunity to prognosticate sulphur capture performance. Though the increasing awareness of the environmental impact of oil shale combustion has led to research on problems related to environment protection, there are yet no successful data about sulphur capture in circulating fluidized bed [9–10]. If sulphur is captured in fluidized bed then on convective surfaces their recarbonation process may take place [11]. The deposition of particles in recarbonation conditions is still not investigated enough.

Aim of the Tests

The goal of this study was to prognosticate the efficiency of Estonian oil shale combustion in circulating fluidized bed.

The present investigation concerns the mechanism of fly ash formation, in particular, that of the fine ash which is most frequently associated with fluidized-bed combustion of Estonian oil shale. Formation and behavior of solid particles up to 100 μm are of great interest as they can cause difficulties in exploitation of heat-exchange surfaces at fluidized-bed combustion [2].

We have also tried to estimate sulphur capture rate in different points of the test rig. Control of sulphur dioxide (SO_2), sulphur trioxide (SO_3) and hydrogen sulphide (H_2S) emissions from different points of the fluidized-bed set-up is one of the main environmental problems.

Experimental

Test Rig

The test rig (Fig. 1) allows burning granulated oil shale both in a bubbling (BB) and in a circulating atmospheric fluidized bed (CFB).

This test rig has two sections. The first one is a fluidized-bed (FB) reactor. It is a ceramic tube with the inner diameter of 100 mm and the height of 4,000 mm, equipped with an electrical heater for creating starting (ignition) conditions. The air is fed to the fluidized bed under the fluidized-bed grate. For feeding fuel to the fluidized bed the screw feeder is used. The floating particles flown out from the fluidized bed are captured in the cyclone and directed through the nozzle and with the screw feeder back to the fluidized bed or gathered in the bunker. The gas scrubbed in the cyclone is directed to the second section, which can be considered convective duct.

The FB reactor and the convective duct are equipped with thermocouples, loopholes for obtaining data on combustion condition, determining flue gas composition, etc., and a special window space for Laser-Doppler-Anemometry (LDA) measurements.

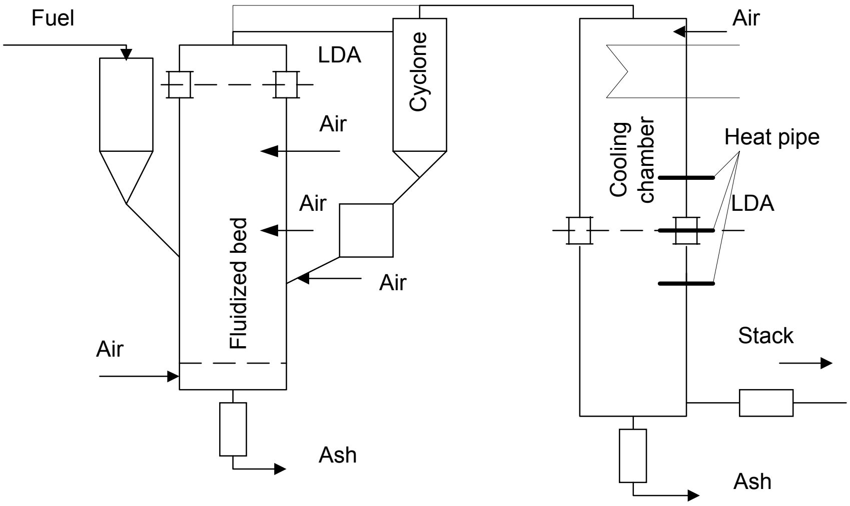


Fig. 1. Flow sheet of the test rig for fluidized-bed combustion of oil shale

The designed thermal load of the test rig is approximately 100 kW (oil shale consumption 40 kg/h). Preliminary tests were performed at capacities about 40–60 kW.

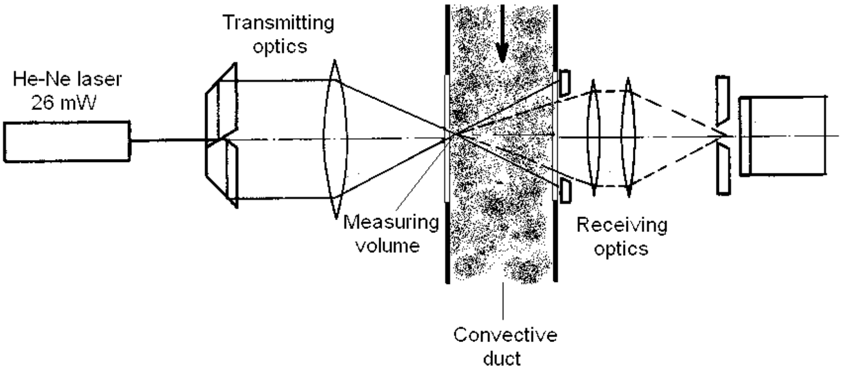


Fig. 2. Schematic diagram of the LDA technique

Technique for Measuring the Velocity and Particle Concentration Fields

The LDA technique adapted to specific conditions of the given experiment was used (Fig. 2). This technique was tested earlier for solving the problems of counting aerosol particles [12]. In our tests it was tuned for recording signals from the particles of the size 4–70 μm. During preliminary tests the LDA measuring volume was located close to the duct wall.

Heat-Exchange Surfaces

Special heat pipe probes [13] were designed for estimating the fouling dynamics of heat-exchange surfaces. The probes were designed as water-filled heat pipes whose working (heat-absorbing) surfaces (with outer diameter of 35 mm) have temperatures on the level of the corresponding surface temperature of an actual boiler superheater (up to 550 °C) or economizer (300–350 °C). That temperature level was reached by placing an insulation layer between heat pipe and probe outer surface. The probes were calibrated in a laboratory electrical furnace.

As a main cooling surface an in-line tube bank consisting of five tubes of a total area of 1.2 m^2 and the tube outer diameter of 25 mm was used. The design of that cooling tube bank allows realizing approximately the same *Re* numbers of the flue gas flow as that in the superheater region of CFB boiler at Narva Power Plant.

Estimation of Sulphur Capture

Sulphur capture in circulating fluidized bed was studied with the aim to find out whether it could be possible to prognosticate the efficiency of this process. For sulphur measurements the technique described in [14–16] was used.

Fuel

Estonian oil shale used was ground in a jaw-breaker to the particle size from 0.5 to 3.5 mm. Its composition is as follows: moisture 1.10 wt.%; ash (at 815 °C) 47.80 wt.%; mineral CO_2 19.09 wt.%; low heat value 11.70 MJ/kg; elementary composition, wt.%: C 31.98, H 3.34, N 0.093, S 1.84, CaO (determined in ash) 49.58.

Test Procedure

Fuel is fed directly into the fluidized bed. Combustion air is fed in two stages: primary air through the nozzle grid at the bottom of the fluidized bed and portions of secondary air at various levels above the fuel feed point to ensure complete burning of fuel.

Experimental conditions were as follows: approximate thermal load of the test rig and thermal load realized during preliminary tests 100 and 40–

60 kW, respectively; mean temperature in the bed 815 °C; excess air coefficient at the outlet of reactor 1.12; size of fuel particles 0.5–3.5 mm; size of ash particles in flue gas 4–70 μm (up to 32%); particle velocity in flue gas 1.8–2.0 m/s; ratio of circulating ash to the ash supplied by fuel 1.7–5.1. Also the content of CO, NO_x and O_2 in flue gas at the exit of FB reactor was measured. The mean fluidized bed temperature during tests was in the range of 815–850 °C.

To estimate sulphur capture, the ash from the fluidized bed, recycle cyclone, heat-exchange surfaces and flue gas was sampled and analyzed. As during preliminary tests there was no possibility to measure directly SO_2 content of the flue gas, an attempt was made to estimate the sulphur capture rate on the basis of sulphur content of the ash samples taken from different points of the equipment (Table 1).

Table 1. **Sulphur Distribution in the Test Rig**

Sulphur form	Value
Sulphur in oil shale, % per ash:	
S_{total}	3.97
$S_{sulphate}$	0.25
Sulphur in FB material, %:	
S_{total}	3.88
$S_{sulphate}$	3.87
Sulphur in circulating material, %:	
S_{total}	3.01
$S_{sulphate}$	2.92
Sulphur in flue gas (probe sample), %:	
S_{total}	3.45
$S_{sulphate}$	3.06
Flue gas volume, Nm^3/kg fuel	4.34
SO_2 concentration in flue gas:	
in mg/Nm^3	0.65
in ppm_{Vol}	2

Results and Discussion

Two preliminary test series were performed: the first one without ash circulation and the second one with circulation of ash at different circulation factors (1.7–5.1, estimated as the ratio of circulating ash to the ash supplied by fuel).

Figure 3 illustrates the isothermity Θ defined by Eq. (2) *versus* different circulation factors:

$$\Theta = T_{aver}/T_{max} \tag{2}$$

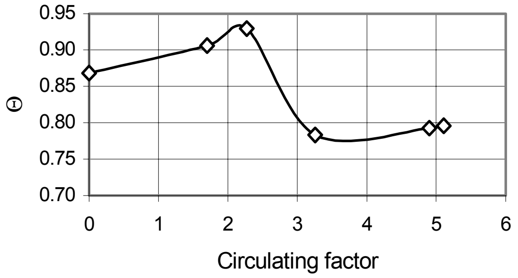
where T_{aver} is average temperature of the medium in the FB reactor, K;

T_{max} is maximum temperature (as a rule for the given case in the reactor entrance), K.

Some variations were noted in temperature distribution lengthwise of the FB reactor depending on the circulation factor of ash.

The temperature field at different circulation factors is presented in Fig. 4. Perhaps, the problems with temperature distribution along the reactor height were induced by low thermal load at preliminary tests, when the temperature at the upper end of the FB reactor decreases sufficiently (especially at higher circulation rates of ash).

Fig. 3. Isothermity factor *vs* circulation rate



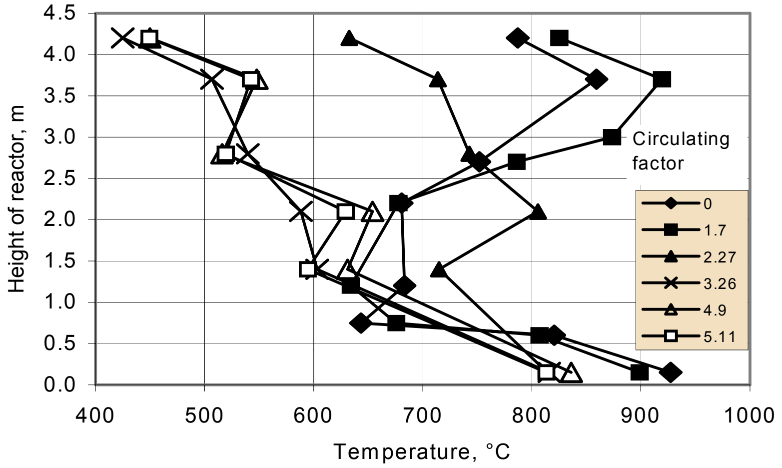


Fig. 4. Temperature distribution lengthwise the FB reactor at different circulating factors

During the measurements the signals from the 4–70-μm particles were recorded. Velocities of those fine particles were in the range of 1.8–2.0 m/s. The counting method was used for determining the concentration of particles in the selected volume of flue gas. The instant time of receiving a signal from each particle in the LDA channel was recorded. We assumed that the particle flow is characterized by the constant mean density, and therefore the results of particle counting obey the Poisson distribution [17, 18]. The technique of data processing consisted of standard procedures [19, 20]. The fixed characteristics of the Poisson distribution allow calculating particle concentration in the selected volume of the flue gas flow for various combustion conditions.

For counting the particle concentration and the known area of the cross-section of the LDA measuring volume, perpendicular to the flue gas flow, the flow density of particle can be calculated. Based on the flow density and mean velocity, the numerical concentration of particles can be estimated. The analysis of the obtained data allows pointing out that the method is quick-response in local transient events quite apart from stable conditions of the fluidized bed.

Formation of hardbound (sintered) ash deposits took place at the exit region of the FB reactor mainly due to the excess air inlet and afterburning of particles at a temperature rising up to 1000 °C at BB conditions. The afterburning at the exit of the FB reactor was confirmed by high CO content of flue gas at that point despite of sufficient excess air. As for the material deposited onto the heat-exchange surfaces during short-term experiments only its little amount could be taken. Nevertheless, this material was thoroughly investigated. The data on fractional analysis of oil shale, bed material and fly ash are given in Fig. 5.

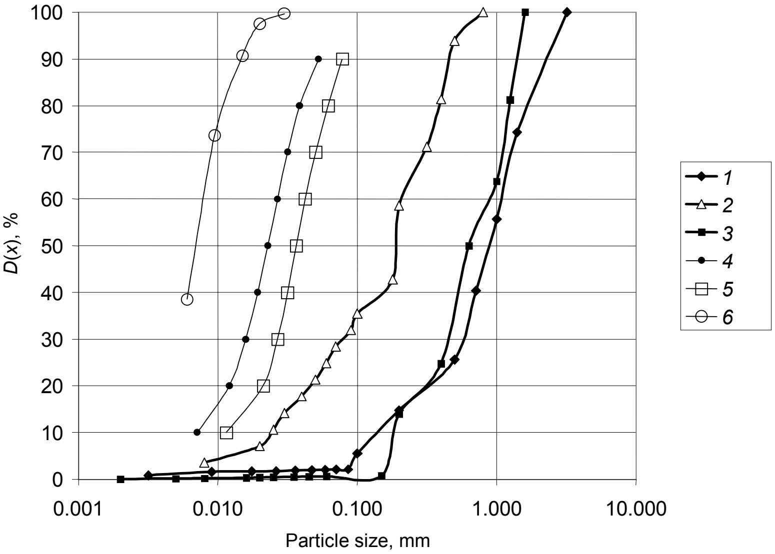


Fig. 5. Fractional analysis: *1* – oil shale, *2* – circulating ash, *3* – bed material, *4* – heat-exchange surfaces, *5* – after-bunker, *6* – fly ash

The comparison of granulometric parameters of fuel and ash samples taken from different characteristic points of test section with those of other tests (Lurgi and Ahlström [21]) is presented in Table 2.

The tests showed that up to 32% of fly ash consists of micron-size particles. Taking into account their tendency to deposit onto heat-exchange surfaces by the molecular, diffusion, thermophoresis and some other forces, intensive fouling of these surfaces may be prognosticated by measuring their concentration in a solid volume-fraction on a local surface.

Table 2. **Comparison of Median Particle Size (mm) of Fuel and Ashes of the Given Test with Other Similar Data on Oil Shale CFB Combustion [21] (*D* is the Reactor Diameter, mm)**

Material	Given test, $D = 100$	Lurgi, $D = 700$ Plant	Ahlström, $D = 600$ Plant
Oil shale	1	2	2
Circulating ash	0.12	0.08	0.105
Fly ash	0.008* (0.025**)	0.015	0.022

* Determined by LDA.
** Ash from the probe in convective duct.

We have found that in the given case (Δ = 4–70 μm) the S_t number describing the possibility of particle deposition on heat-exchange surfaces equaled 0.16, while $S_{t_{cr}}$ was 0.12. As $S_t > S_{t_{cr}}$ for the given situation (the cross-flow probe with outer diameter of 35 mm), the micron-size particles are settling on the heat-exchange surfaces of the circulating fluidized bed test rig. It was really observed in the given temperature conditions. The ash deposits on the cross-flow probes after a 3–4-h test period can be characterized as typical weakly-bound easily-removable deposits without any signs of sintering (Fig. 6). A noticeable amount of deposits was formed in BB (without circulation) conditions. Here it must be noted that during preliminary tests the temperature of typical flue gas in a boiler convective part was not reached. Due to that it is difficult to speak about a true nature of the phenomena.


Result of fouling at circulation rate of 1.7. Deposited ash is fine, median particle size 15–20 μm


Result of fouling at circulation rate of 4.9


Result of fouling of cross-flow probe in BB conditions. The intensity of fouling is higher and ash coarser

Fig. 6. The character of ash deposits on cross-flow probe in convective duct at different conditions used

As for the sulphur capture estimation, these investigations were carried out in EERI with the purpose to study the basic mechanism of this process and recommend thermal conditions for environmentally friendly utilization of oil shale [14–16]. As noted above, the sulphur capture estimation based here on the differences in sulphur content of ash samples taken from different places of the test rig (Fig. 7).

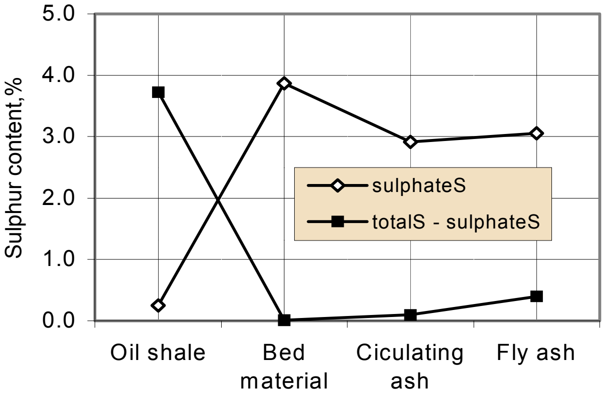


Fig. 7. Sulphur content of oil shale and different ashes

At oil shale thermal processing (including its combustion in FB) sulphur compounds, except sulphates, will decompose and gaseous sulphur compounds SO_3 and SO_2 will react with free CaO. The increasing sulphate content of the ash passing the FB confirms sulfur capture.

Basic sulphides present in fuels and their possible reactions are thoroughly described [9, 10, 14–16]. Yet it should be noted that various compounds of sulphur behave differently at thermal processing: ones are transformed and remain in ashes, others are decomposed and stay in flue gas as SO_2. The character of the final products depends on thermal processing conditions: whether it is carried out in oxidizing or in reducing surroundings.

The basic component in the oil shale mineral part responsible for sulphur capture is calcium present in carbonates. As for oil shale, it is necessary to take into account that calcium is in the mineral part, and thus it is possible to analyze the weight ratio of sulphur to calcium in fuel.

The range of sulphur fixation by fly ashes depends both on temperature and on time of contacting hot gases with fine solid particles. Table 1 lists the data obtained during the experiments carried out in circulating fluidized bed at 815 ºC. One can note that the sulphur capture level is quite high. By the approximate estimation SO_2 content of flue gas was at tests on the level of several ppm, which is quite a low content compared with the data of PF combustion of Estonian oil shale (1,500 Nm^3/kg [22]).

The preliminary tests of burning Estonian oil shale in a small-scale circulating fluidized-bed unit have shown that the conditions in the FB reactor and flue gas velocities both can play a certain role in fouling of convective surfaces and in sulphur capture.

Conclusions

The preliminary test results allow formulating the following conclusions:

1. The laboratory test rig designed for investigating oil shale behavior in circulating fluidized bed has passed preliminary tests which have demonstrated its applicability to study the behavior of heat-exchange surfaces and sulphur capture in complex conditions. The experimental technique and data processing may be realized in actual complex conditions of burning Estonian oil shale in circulating fluidized bed.
2. The LDA method is suitable for measuring the concentration of micron-size particles near the heat-exchange surfaces of a high-temperature fluidized-bed reactor. It helps to prognosticate the behavior of fine solid particles and transient events in fluidized-bed reactor burning Estonian oil shale.
3. The study of the samples taken from characteristic points of the test rig allowed estimating of possible fouling of heat-exchange surfaces.
4. For elucidating the mechanism of sulphur capture in the fluidized bed the data on the formation of sulphides from oil shale particles in the FB reactor and along the convective duct are required. The influence of temperature, bed height and behavior of solid particles on the rate of sulphur capture by ash particles are of the practical interest.

These are the first results of the experiments carried out to determine the importance of combustion conditions on the size and character of fly ash particles, as well as their influence on fouling of heat-exchange surfaces.

Further experiments on the described test rig are needed to get more data for creating a mathematical model for prognosticating the behavior of oil shale in an industrial-scale reactor with circulating fluidized bed.

Acknowledgements

This research was supported by Estonian Science Foundation (Grant No. 5646). We thank Msc. Medhat Hussainov for his co-operation. Our special thanks are due to Dr. Ülo Rudi and Mr. Ago Hio for their efforts and help enabling us to carry out the present investigation.

REFERENCES

1. Oil Shale Perspectives within Energy Production (First Inception Report) – LURGI, March 1996.
2. *Prikk, A., Arro, H.* Circulating fluidized-bed combustion. The technology exact for Estonian oil shale // Oil Shale. 1997. Vol. 14, No. 3 Special. P. 209–214.
3. *Rosner, D.* Transport Processes in Chemically Reacting Flow Systems. – Butterworths, 1986.

4. *Tsuji, Y., Morikawa, Y., Shiomi, H.* LDV measurements of an air-solid two-phase flow in a vertical pipe // J. Fluid Mech. 1984. V. 139. P. 417–434.
5. *Lee, S.L., Durst, F.* On the motion of particles in turbulent duct flow // Intern. J. Multiphase Flow. 1982. V. 8, No. 2. P. 125–146.
6. *Hussainov, M., Kartushinsky, A., Mulgi, A., Shcheglov, I., Tisler, S.* Some results of investigation of solid particles distribution in the vicinity of various shapes in two-phase flow and sedimentation on their surfaces // Israel, Haifa, Technion City: Proc. 27th Israel Conf. on Mechanical Engineering. May 19–20 1998. P. 356–358.
7. *Ots, A.* Thermal Engineering Department of Tallinn Technical University // Oil Shale. 1997. Vol. 14, No. 3 Special. P. 195–208.
8. *Ots, A., Tiikma, T.* On the fouling mechanism of non-bounded oil shale ash deposits on boiler heat transfer surfaces // *Ibid*. 2003. Vol. 20, No. 1. P. 3–13.
9. *Lyngfelt, A*., *Leckner, B.* Sulphur capture in circulating fluidized-bed boilers: can the efficiency be predicted? // Chem. Eng. Sci. 1999. Vol. 54. P. 5573–5584.
10. *Anthony, E.J., Granatstein, D.L.* Sulfation phenomena in fluidized-bed combustion systems // Progress in Energy and Combustion Science. 2001. Vol. 27. P. 215–236.
11. *Yoffe, O. et al.* The chemistry and mineralogy of the Negev oil shale ashes // Fuel. 2002. Vol. 81. P. 1101–1117.
12. *Dushenko, V.*, *Hussainov, M., Shcheglov I., Tiikma, T.* Formation of aerosol particles by low-frequency sound-stimulated combustion // Proc. Estonian Acad. Sci. Eng. 2003. V. 9, No. 2. P. 137–147.
13. *Vrager, A.*, *Tiikma, T.* Properties of thermal probes with heat pipe cooling system // Lithuania, Kaunas: Proc. 4th Baltic Heat Transfer Conf. August 25–27, 2003. P. 587–594.
14. *Uuesoo, R.*, *Kallaste, E.* Investigation of the Process of Sulphur and Chlorine Capture in Fluidized Bed at Preliminary Gasification of the Estonian Oil Shale : Final report (VINITI No. Б 175 836). – Tallinn : The Estonian Academy of Sciences, Institute of Thermophysics and Electrophysics (since 1996 – EERI), 1971 [in Russian].
15. *Uuesoo, R.*, *Kallaste, E.* Investigation of the Reactions of Sulphides and Chlorides at Burning High-Ash Fuels : Final report (VINITI No. Б 606 925). – Tallinn, The Estonian Academy of Sciences, Institute of Thermophysics and Electrophysics (since 1996 – EERI), 1975 [in Russian].
16. *Kallaste, E.* Distribution of Sulfur at Thermal Preparation of Oil Shale in Fluidized-Bed Test Rig : Dissertation. – Tallinn : Tallinn Technical University, 1978 [in Russian].
17. *Tyurin, J.N., Makarov, A.A.* Statistical Analysis of Data with a Computer (V.E. Figurnov, ed.). – Moscow : *Infra*, 1998 [in Russian].
18. *Dubnishchev, Yu.N., Rinkevichyus, B.S.* Technique of Laser Doppler Anemometry. – Moscow : Nauka, 1982 [in Russian].
19. *Klochkov, V.P., Kozlov, L.F., Potykevich, I.V., Soskin, M.S.* Handbook of Laser Anemometry. – Kiev : *Naukova Dumka*, 1985 [in Russian].
20. *Shchenck, H.Jr.* Theories of Engineering Experimentation. – New York : McGraw-Hill, 1971.
21. *Arro, H., Prikk, A., Kasemetsa, J.* Grain composition and corrosive activity of ash from CFB oil shale boiler // Oil Shale. 1997. Vol. 14, No. 3 Special. P. 225–235.
22. *Arro H., Prikk A., Pihu T.* Fouling and corrosion of heat transfer surfaces of FB boilers burning Estonian oil shale // Oil Shale. 2001. Vol. 18, No. 3. P. 193–202.

Presented by A. Ots
Received December 5, 2003