RESEARCH OF THE FLUIDIZED BED COMBUSTION IN THE LABORATORY FOR THERMAL ENGINEERING AND ENERGY
Part B: Achievements in Technology Implementation

by

Borislav D. GRUBOR[*], **Dragoljub V. DAKIĆ, Stevan Dj. NEMODA, Milica R. MLADENOVIĆ, Milijana J. PAPRIKA, and Simeon N. OKA**

Laboratory for Thermal Engineering and Energy, Vinca Institute of Nuclear Sciences,
University of Belgrade, Belgrade, Serbia

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Paper gives a review of the most important results of extensive and wide-ranging research program on R&D of fluidized bed combustion technology in the Laboratory for Thermal Engineering and Energy of the VINCA Institute of Nuclear Sciences. Paper presents detailed overview of R&D activities from the beginning in the second half of the 1970's up to present days. These activities encompass applied research achievements in the field of characterization of limestones and bed agglomeration and sintering and modeling of overall processes during fluidized bed combustion, all of which have facilitated the R&D of the fluidized bed combustion technology. Attention is also given to steady-state combustion testing of a wide-range of fuels (coals, liquid fuels, biomass, waste solid and liquid materials, etc.) in our fluidized bed combustor and development of original methodology for testing the suitability of fuels for fluidized bed combustion, as well as specific achievements in the area of technology application in Serbia.

Key words: *fluidized bed combustion, applied research, limestone characterization, bed agglomeration, mathematical modeling, technology development*

Introduction

In the second half of 1970's in former Yugoslavia there was a considerable interest for new combustion technologies which could enable wider use of local fuels such as low quality coals, biomass and wastes. This interest was a direct consequence of the oil crisis at that time. In the Vinca Institute of Nuclear Sciences, Laboratory for Thermal Engineering and Energy (in further text: LTEE) a program for R&D of fluidized bed combustion (FBC) technology was started since our analysis led to the conclusion that FBC technology was the most promising one for domestic solid fuels.

The aim of this paper is to show the FBC R&D efforts in LTEE and our direct involvement in the implementation of FBC technology in former Yugoslavia. These activities encompass some specific applied research results, modeling overall processes during FBC and, of course, fuel testing as well as our direct involvement in design and implementation endeavors.

In spite of the fact that former Yugoslavia was not a highly developed country (not even having a producer of electricity generating boilers) and that the R&D activities were modestly funded, LTEE has become one of the most highly respected FBC R&D centers for both fundamental research and technology implementation, certainly the most important one in West Balkans, and perhaps in South-East Europe.

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[*] Corresponding author, e-mail: grubor@vinca.rs

Applied research in realistic conditions

Characterization of limestones

The use of limestone as a SO_2 sorbent in fluidized bed combustion of coal is a well-known practice worldwide. For design purposes it is necessary to know the properties of limestones that determine their sulfur capture potential, and in turn the design of limestone storage and feeding equipment. The investigations in LTEE consisted of characterization of 9 different limestones in former Yugoslavia, defining mechanical and flow characteristics of limestones, porosity before and after calcination, and the sulfation degree of each limestone [1, 2]. The experimental procedures were based on the results of investigations of prominent authors in this area at that time [3-5].

The kinetics of the process of calcination was studied, monitoring the change in limestone batch mass upon introduction in a pre-heated laboratory oven. Varied were the particle size classes (0.4-0.5 mm, 0.8-1.25 mm, 1.25-2.0 mm, 2.5-3.15 mm) and the temperature at which calcination took place (650 °C, 700 °C, 750 °C, 800 °C, 850 °C). It was noted that the limestone type practically had no influence on the duration of calcination, but that limestone particle size and calcination temperature have a pronounced influence on the process [1].

The porosity of limestone samples before and after the process of calcination was also determined using low temperature static gas adsorption method. Initial surface area of pores ranged from 0.2-5.6 m^2/g while after calcination the obtained values were in the range 6.6-15.7 m^2/g. But the increase in surface area of pores varied drastically for various limestones (from 1.5 to 70 times increase). Initial total pore volume ranged from 0.001-0.028 cm^3/g and the obtained values after calcination were 0.025-0.2 cm^3/g. For various limestones the increase in total pore volume before and after calcination was from 1.5-150 times fold.

For investigation of the reaction between limestone and SO_2 (process of sulfation), a method was chosen for which a reactor with inner diameter of 39 mm with fluidized bed of sand is used. Fluidization gas simulated a typical flue gas mixture obtained during combustion of our lignites. In a pre-heated fluidized bed, at a desired temperature, a batch of limestone is introduced and the change in the outlet gas composition is monitored [2].

Analysis of limestone behavior was made based on the values of the degree of sulfation which is defined as the used up amount of both $CaCO_3$ and $MgCO_3$ in relation to the initial amount in the batch of limestone:

$$X(\tau) = \frac{S_{\text{tot}}(\tau)}{\text{CaCO}_3 + \text{MgCO}_3} \cdot 100\ [\%]$$

$S_{\text{tot}}(\tau)$ (mol So_2) - total SO_2 absorbed by limestone up to time, τ
$CaCO_3 + MgCO_3$ (mol) - the $CaCO_3 + MgCO_3$ content in limestone

On fig. 1 are shown the characteristic change of the degree of sulfation in time for all 9 types of limestone (granulation 0.4-0.5 mm), at 850 °C bed temperature. It can be seen that initially sulfation degree increases rapidly and then asymptotically reaches a maximum value as maximum conversions are attained (after 20-30 minute depending on limestone type). The same figure shows the influence of bed temperature on sulfation degree (maximum values). It can be seen that for majority of limestone types significant increase of SO_2 retention is noted when bed temperature is higher than 820-825 °C reaching maximum values (10-23%) at 850-860 °C.

The differences between limestone types and especially the behavior of limestone No.6 could not be fully explained. Neither the chemical composition nor the porosity before or after calcination could account for differences in obtained sulfation degrees for different limestone types. This only emphasizes the necessity of experimental determination of the performance of limestones in fluidized bed conditions.

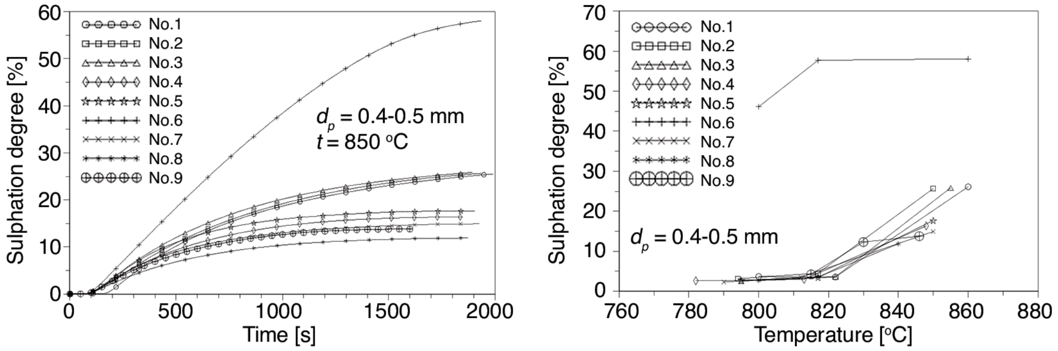

Figure 1. Degree of sulfation *vs.* time and temperature for different limestones [2]

Bed agglomeration

During the early R&D phase (of fluidized bed furnaces), the combustion tests with various biomasses in laboratory fluidized bed facilities showed very promising results. With combustion temperatures above 800 °C and adequate split of combustion air into primary and secondary, for each type of biomass, the measured combustion efficiencies were above 97% and the CO emissions were generally below 500 ppm. Apart from minor fuel manipulation difficulties, no drawbacks of the use of fluidized bed technology for combustion of biomass could be noticed.

However, all built industrial FBC hot-gas generators, burning corn cob, experienced severe difficulties in operation [6, 7], *i.e.* agglomeration and sintering problems causing shutdown and requiring complicated replacement procedures. Thus, the operation of these fluidized bed furnaces was possible only by lowering the combustion temperature below 700 °C. This only somewhat reduced the mentioned problems but on the other hand resulted in unacceptable CO emission levels.

In order to overcome these difficulties, an experimental program has been organized to study the relevant phenomena that lead to agglomeration and sintering problems and to point out possible solutions. With this aim in mind, long term steady-state combustion of corn cobs at 820 °C in an experimental 200 kW fluidized bed furnace was organized, taking samples of bed material every 12 hours for chemical analysis. After several days the operation had to be aborted because of severe agglomeration and sintering manifestations. The testing was repeated at a lower temperature of 750 °C with the same outcome but only after a longer period of operation.

Analysis of data showed that in two regimes approximatly 43% and 39% of the alkalines, fed with corn cobs, was deposited in the bed, while for Fe these values were approximatly 56% and 49%. This indicates that at higher temperatures (regime I) both Si (in bed) and Fe (in corn cob) react more readily with the alkalines and that Fe in both regimes reacted to a greater extent (relative to the amount fed) than Si.

These experiments showed that coal ash reacts with the inert solid material (in our case silica sand), forming mixed oxides and the corresponding eutectic mixtures with low melting temperatures. The most probable reactions (undesirable from agglomeration point of view) that take place are the ones by which the alkaline oxides (salts) in ash react with Si forming the following mixed oxides:

$$2SiO_2 + Na_2CO_3 \rightarrow Na_2O \cdot 2SiO_2 + Co_2$$
$$4SiO_2 + K_2CO_3 \rightarrow K_2O \cdot 4SiO_2 + Co_2$$

i.e. eutectic mixtures with melting points (874 °C and 764 °C respectively) which are lower than for the individual components. Data published in [8] indicated that transitional metals (in ash Fe is dominant) also readily react with alkalines as follows (with X meaning Na or K):

$$Fe_2O_3 + X_2O \rightarrow X_2Fe_2O_4$$
$$Fe_2O_3 + X_2CO_3 \rightarrow X_2Fe_2O_4 + Co_2$$

by which mixed oxides and eutectic mixtures are formed with a minimum melting temperature of 1135 °C. Such reactions are very desirable since they would result in less alkalines available to react with Si. From the chemical composition in ashes of the biomasses it cannot be concluded that this phenomena would lead to greater problems than those encountered with coals. We were able to justify the differences only after the following rational, *i.e.* after the analysis involving the parameters which we named as the specific Fe and Si (molar) content in ash:

$$\frac{Fe_2O_3}{Na_2O + K_2O}\left[\frac{mol}{mol}\right] \quad \text{and} \quad \frac{SiO_2}{2Na_2O + 4K_2O}\left[\frac{mol}{mol}\right]$$

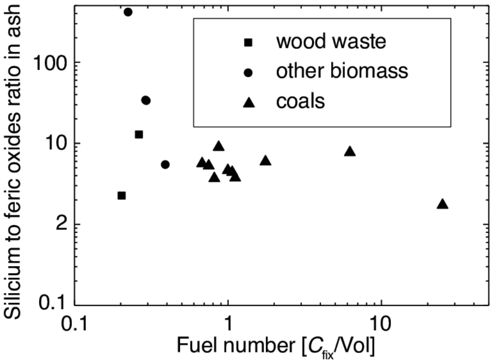

Figure 2. Specific Si to Fe ratio for the fuels tested [6]

Namely, we found that the tendency for agglomeration is more pronounced in those fuels that have higher specific Si to Fe ratio, fig. 2.

Clearly, silica sand is not an acceptable bed material for combustion of biomass with large alkaline content in ash (especially for those which are fertilized). Two materials were chosen for further investigations: corundum (Al_2O_3) and ferric oxide (Fe_2O_3), because of their availability and relatively low cost. Our choice of materials was also based on one patent application that we found in literature [9]. Two types of tests were used: heating-up of premixed samples of these materials and alkaline oxides, and feeding of alkaline solutions into a fluidized bed reactor with these materials as bed materials. These tests clearly showed that ferric oxide (Fe_2O_3) is more resilient and that agglomeration with this as inert material may be expected only above 900 °C.

The full scale industrial verification tests were performed on a 4 MWth FBC hot gas generator furnace (burning corn cob) in the corn seed center in Subotica. Silica sand was replaced with pure hematite and the furnace performance was monitored during normal operation. The improvement in the operation of this fluidized bed furnace in 1995, after replacement of silica sand with hematite, was remarkable [10]. The operation at 715 °C was stable for over 4500 hours (two full grain drying seasons), the CO concentration being less than 200 ppm and the overall furnace efficiency increased to nearly 80 %. The operation of the furnace was stopped several times for inspection purposes and no fouling of furnace walls, furnace exit and flue-gas/air heat exchanger was noticed.

Fuel testing in FBC furnace

One of the main features of the FBC boilers/furnaces is flexibility to the origin and quality of fuels. By this feature, FBC differs greatly from the classical grate firing boilers and pulverized coal combustion boilers. From the phenomenological point of view, large amount of data and experience in the world have led to fairly reliable guidelines on the influence of various fuel characteristics on behavior during FBC as well as on boiler/furnace design [11-17]. It should be noted that these guidelines are only of a qualitative nature and cannot be taken as a reliable basis for any specific boiler/furnace design, but perhaps only as a guide for further needed investigations for any particular fuel and application. This is especially true when considering some low rank coal or waste fuel, the very kind of fuels that FBC technology was considered for. Therefore, in early 1980's we have started to build-up our experimental basis.

Firstly we built several small test rigs experimental studies of specific processes such as coal particle devolatilization, char particle combustion, coal particle fragmentation, limestone characterization, sulfur self retention by ash, *etc*. The results of these investigations were shown in previous sections.

For obtaining the relevant data for boiler/furnace design we built an experimental 200 kW fluidized bed furnace. The unit has a cross-section of 200 × 200 mm and the height of the freeboard is appr. 1.5 m and a more detailed description is given in [18-20]. The fuel may be fed both in-bed and over-bed, and the auxiliary equipment and measurements of pressure, temperature and gas composition at various points, as well as particle sampling enabled us to define the following:

- Heat and mass balance of all species.
- Temperature distribution in freeboard.
- Heat released in bed and in the freeboard.
- Amount and size distribution of ash retained in bed and of fly ash.
- Concentration profiles of O_2, CO_2, CO, SO_2 and NO_x in the freeboard.
- Combustion efficiency.
- Sulfur capture degree with various Ca/S ratios.
- Influence of primary to secondary air ratio.
- Transient behavior during the change from one to another regime, *etc*.

We have tested many fuels in this unit: various types of coals [18-21], biomasses [22], as well as liquid and waste fuels such as coal rejects and paper waste [23-25]. Several types of lignites were also tested with the aim to determine their suitability for use in CFBC units [26]. Specific attention was given to devolatilization and pyrolysis of coal [27], resulting also in an M. Sc. thesis [28], since the content of volatiles in coals from our region is significantly higher than in coals reported in literature.

The gained experimental experience and the results of investigations enabled acquiring a large data base for coals and a formulation of a methodology of fuel investigation which was best documented in [29] and will be shortly presented in later text. Our broad conclusions and guidelines may be summarized:

- *Low rank coals* (lignites, high-volatile bituminous coals) usually need a simple boiler concept: no pretreatment of coal, over-bed feeding of as received coal, no recycling system, periodical bottom ash discharge, small or none limestone addition, low bed height, high freeboard, probably secondary air. Start-up temperatures are lower than 600 °C, and consequently a smaller start-up chamber or burner is needed. The amount of fly ash is high and a large capacity of flue gas cleaning system is expected, as well as a relatively higher amount of fresh sand addition.
- *High rank coals* (low volatile bituminous, anthracite) need in-bed pneumatic feeding, coal grinding and sometimes coal drying, recycling system, bed height control, continuous bottom bed discharge, and a deep bed. Start-up temperatures are higher, up to 800 °C, and the start-up chamber/burner must be large, with long start-up procedure.

- *Separated and washed coals* will not need fly ash recycling system, and bottom ash continuous discharge system.
- *As mined coals* will probably have large amount of stones, and large coal particles (larger than 50 mm). Bottom ash continuous discharge system will be necessary, and coal pre-grinding.
- *Fuels with large moisture content* (biomass) will not need immersed heat transfer surfaces, and probably will need lining of furnace walls.
- *High volatile, small sized coals (biomass)* have to be fed under the bed, within deep bed, and with secondary air supply.
- *High volatile coals, with large particles (lignites)* can be fed over bed. High volatile coals presumably are highly reactive coals, and recycling system is not necessary. Start-up temperatures are low.
- *High ash content* means permanent bed height control, continuous bottom ash discharge, coal pre-grinding, and in-bed feeding. No fresh sand is necessary. Opposite is expected for low ash content as well as for washed and dried coals.
- *Fuels with large content of fine particles* (<1 mm) have to be burned in boilers with deep bed, with in-bed feeding and recycling system.
- *Low rank, high volatile coals* will have higher combustion efficiency (95-98%), even without recycling system.
- *High sulfur coals* will need limestone addition, and appropriate storage and feeding system. Amount of limestone added depends on limestone characteristics and emission standards.
- *Ashes with high alkali metal content* and low sintering temperature have a tendency to agglomerate with silica sand. Another type of inert bed material is necessary (especially for certain kinds of biomass). Risk of bed agglomeration has to be considered.

In the early 2000's, a new 500 kW experimental unit was built, shown on fig. 3, with similar capabilities as the 200 kW unit, but which is of a circular cross-section. Several fuels have been tested in this new unit such as hazelnut shells [22], liquid waste fuels [23] and paper waste [25]. Specific investigations have also resulted in a Ph. D. thesis in the area of decomposition of high density liquid fuels in fluidized bed [30].

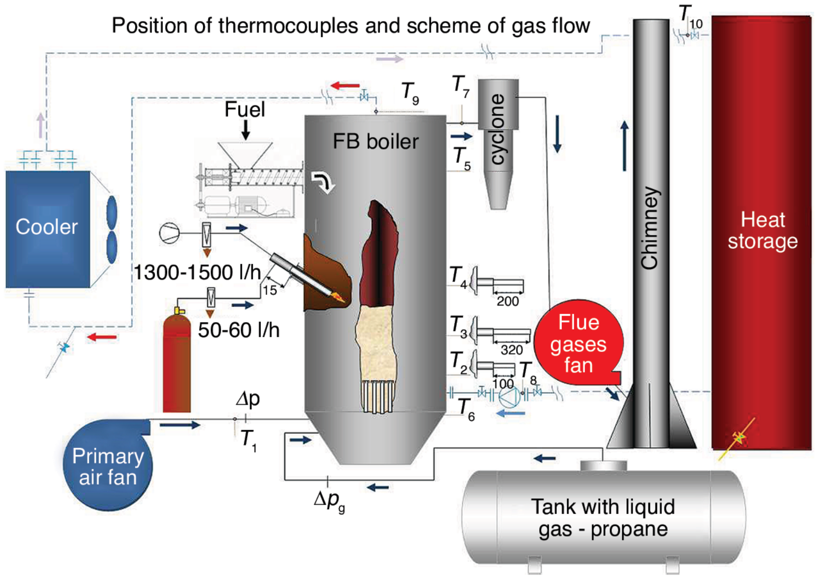

Figure 3. A 500 kW fluidized bed furnace for steady-state combustion tests

Modeling of overall processes during FBC

The modeling of the overall processes during FBC of coal has started in LTEE as a result of our involvement in the IEA agreement in the area of Atmospheric Fluidized Bed Combustion. Namely, an IEA-FBC model was adopted as an improved and extended version of the CANMET, based on the work in [31-33].

Besides the usual activities related to the co-operation in the modeling group of the Agreement [34], we came to the conclusion that the IEA-FBC model should be significantly modified to suit our low grade lignites. The results of our efforts may be seen from the papers in which we published the results we obtained comparing our test results with the model [35, 36], introduction/suggestion of new modules [37, 38], especially in the area of char combustion, volatile matter distribution, over-bed fuel feeding, *etc*. All of this research resulted also in one Ph. D. thesis [39] by Dr. Borislav Grubor who led the modeling activities in LTEE.

The approach to overall modeling is based on the two-phase model for bubbling fluidization. A representation of one bed slice is given on fig. 4 and it can be seen that various mass streams are grouped into six main groups. Mass balances for each molecular species can be estimated by consideration of each of the efflux processes involved. The summation of the mass balances over all molecular species gives the total mass balances.

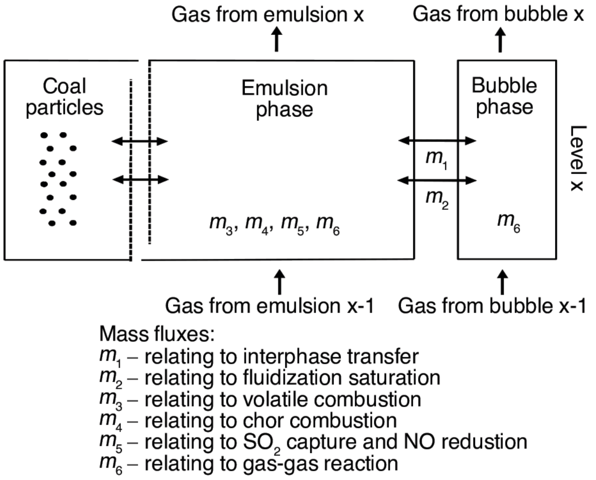

Figure 4. Schematic diagram of a level in the IEA-FBC model [34]

The various processes that coal particles undergo when fed over-bed, before reaching the bed surface, are schematically shown on fig. 5. Depending mainly on their size and other characteristics, certain class sizes of coal will undergo some or all of the stated processes: heating-up and drying, devolatilization and char combustion up to a certain degree. In addition, particle fragmentation, attrition and elutriation are also taken into account. All of these processes influence the heat and mass balances in the freeboard zone, and this is taken care by introducing additional heat and mass sinks and sources in the freeboard calculations.

The algorithm of the model is as follows. The change in coal particles that reach the bed are determined first, if the coal is fed over-bed. The bed fluid dynamics is treated next to determine the amount of gas in the emulsion and bubble phase, distribution of bubbles in the bed and the bed expansion. The mixing pattern of char particles in the bed is determined and the heat transfer coefficients (for in-bed tubes and walls) are calculated using well known empirical correlations. Next, the coal particle size distribution after primary fragmentation is determined (that occurs during devolatilization) using our modeling approach. Also, various time scales are evaluated (duration of drying, devolatilization and char combustion processes, as well as turn-over times for all size fractions).

At this point, iterative calculations begin with the aim to determine the coal feed rate that satisfies energy and mass balances. For the current value of coal feed rate the volatile matter and char particles distribution is determined. For the volatiles this is done by comparing the duration of devolatilization process and the turn-over times for all size fractions and denoting the appropriate amounts of volatile matter locally at the feed point, uniformly over the bed and above the bed. Distribution of char particles in the bed is determined using the mixing coefficients calculated above. Based on the determined volatile and char distribution, the formation and reduction of various molecular species is estimated grouping them into the above mentioned six main groups of fluxes.

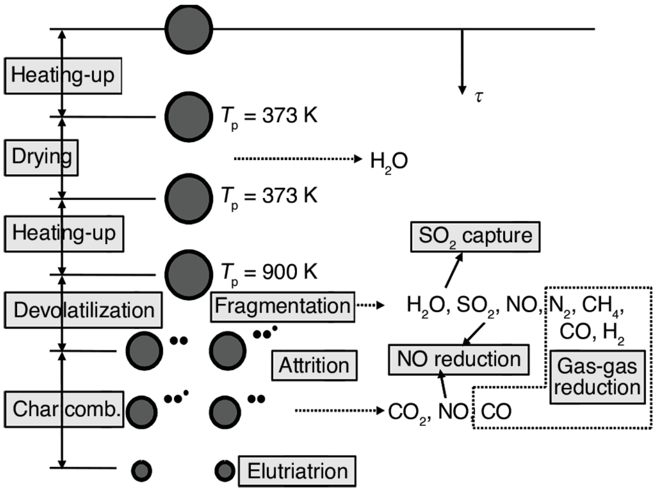

Figure 5. Treatment of over-bed fed coal particles before reaching the FB [38]; τ – the time, T_p – particle temperature

All of the previous calculations are done locally (for each bed slice) and then the mass balances are integrated throughout the bed height. The procedure is repeated till convergence criteria for all molecular species are satisfied. In the freeboard all of the previous processes are practically treated in a similar way adding various fluxes from the coal particles fed over-bed (if that is the case) to the corresponding ones leaving the bed.

On fig. 6 experimental data for combustion efficiency are plotted as a function of the ratio of volatile matter and fixed carbon content (the inverse value of the so called fuel number). Only the regimes without re-circulation are taken into account. As can be seen, combustion efficiency decreases as the ratio of Vol/C_{fix} decreases, *i.e.* as the coal quality increases. This is mostly a consequence of higher attrition rates and slower kinetics of combustion of high rank coals which results in higher unburnt carbon content in fly ash. It can also be seen that the model predicts smaller combustion efficiencies in the case of over-bed feeding. Model's ability to predict flue gas concentrations can be seen in fig. 7. Nearly all experimental results for various coals and combustion conditions are within 15% of the model predictions.

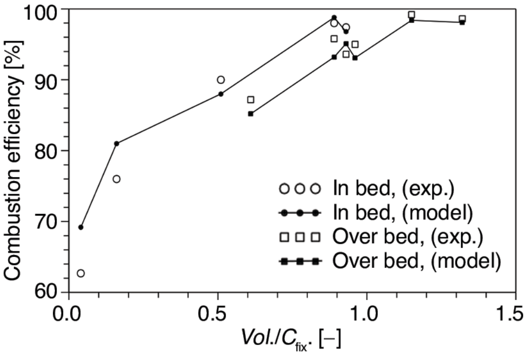

Figure 6. Combustion efficiency; measured values and model predictions [39]

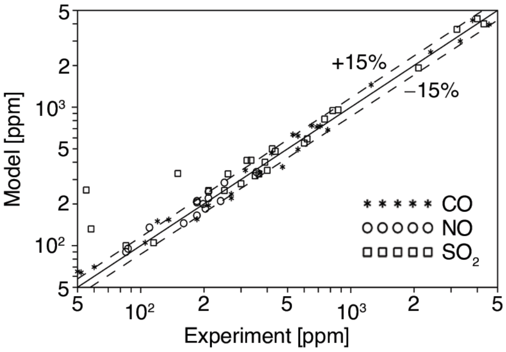

Figure 7. Gas emissions; comparison between measured values and model predictions [39]

Application achievements

As stated previously, the main aim of initial R&D of FBC technology was to obtain as quickly as possible the necessary know-how and experimental data to be able to assist the domestic industry in their efforts to start-up their own development of FBC furnaces and boilers. From literature survey it was clear that our research program should enable us to assist the industry in following aspects:

- Is it necessary to previously prepare the coal (separation, drying and grinding)?
- Could the coal be fed over-bed or must it be fed in-bed?
- Is it necessary to introduce secondary air, and to what extent?
- Is it necessary to re-circulate unburned char particles?
- Is it necessary to build a system for discharging the excess inert bed material, and will this system operate continuously, or intermittently?
- What start-up temperature has to be chosen, what is the size of the start-up chamber and how to organize start-up?
- Are the immersed heat exchangers necessary, and how to distribute heat removal?
- Is it necessary to feed the limestone in the furnace, and what is the optimal Ca/S ratio?

From literature data obtained in laboratory and pilot-plants in the developed countries, and by evaluating our own experimental experience, optimal values of many regime parameters of BFBC boilers, irrespective of fuel characteristics, could be determined. For example: fluidization velocity (1-2 m/s), excess air (1.2-1.4), bed temperature (800-850 °C), inert material (quarts sand) particle size (1-1.5 mm).

Other parameters and design considerations greatly depend on fuel characteristics and cannot be determined only on the basis of conventional proximate and ultimate fuel analysis. At the very beginning of the R&D FBC program it was concluded that it is necessary to develop a special methodology for fuel testing, by which it would be possible to find out the influence of fuel characteristics on the boiler concept and design, and which would give answers on the afore mentioned questions. After several years of investigations, this was accomplished and published in a number of papers [20, 40, 41].

As the number of fuels tested grew, it was possible to form a data base which incorporated our as well literature data which could facilitate the evaluation of the next fuel to be tested. More than 25 coals from Serbia and former Yugoslavia were investigated, having the following range of characteristic:

– Granulation:	0-50 mm
– Moisture (as received)	up to 55% (biomass up to 60%)
– Ash content (dry basis)	up to 75%
– Volatile matter content (dry basis)	up to 45% (biomass up to 75%)
– Ratio of Cfix/Vol	0.76-25
– Lower heating value	4.7-35 MJ/kg

Our fuel investigations may be divided into three phases. In Phase I the fuel is characterized using standard procedures which also include determining sintering temperatures and sink-float tests. Phase II is comprised of investigating specific processes in fluidized bed conditions, as described before (primary fragmentation, sintering temperatures in fluidized bed, ignition temperature in fluidized bed, start-up temperature for fuel feeding in fluidized bed, combustion kinetic tests in fluidized bed, sulfur self-retention in coal ash, sulfation degree of chosen limestone). In Phase III the combustion tests are performed in steady-state fluidized bed conditions in our 200 kW experimental unit, as described earlier.

The analysis of the test results enabled us to come to the following conclusions on the suitability of lignites from this region for BFB combustion [20, 26, 40, 41]:

- High combustion efficiency can be achieved (98-99% and more) without fly ash recirculation for coals having the ratio of the fixed carbon to volatile matter content ($C_{\text{fix}} / VM_{\text{O}}$) less than approximately 1 (younger coals). For higher ratios of these parameters (older, higher quality coals) the combustion efficiency drops rapidly reaching values less than 80% for the ratios of $C_{\text{fix}} / VM_{\text{O}}$ higher than 2. Thus, for such coals fly ash recirculation is absolutely necessary.
- Lignites can burn efficiently without grinding and over-bed feeding may be used, leading to low investment and exploitation costs. This is a consequence of high reactivity due to the high char particle porosity after devolatilization.
- For lignites the starting temperature can be as low as 350-400 °C. This means that the start-up chamber can be very small, and the consumption of the liquid or gaseous fuel during the start-up procedure relatively low. Our methodology defines also a procedure for determination of starting temperature on the basis of the combustion rate.
- During combustion of coals with high volatile content (lignites) even 20-40% of the heat is generated in the freeboard by combustion of volatile matter and fine particles. Distribution and amount of heat removal surfaces in the bed and in the freeboard depend on this heat distribution. Due to this fact, especially when burning lignites as mined (with high moisture content), immersed heat exchangers may not be necessary. Small bed heights can be chosen and as a consequence primary air fan with smaller pressure head may be used. The down side is the fact that smaller specific heat generation per unit of the cross section area is obtained in case of coals with high volatile content, around 1 MWt /m^2 instead of 2 MWt /m^2 which can be achieved with high-rank coals.
- Large amount of ash is elutriated from the furnace during BFBC lignite combustion. Especially when burning separated and washed lignite, almost all ash is elutriated by flue gases from the furnace. A tendency was observed that the part of the coal ash that remains in the bed increases with the increase of contents of SiO_2 and Al_2O_3, although there are always exceptions and testing cannot be avoided.

The achievements of LTEE in the area of application of FBC technology may be seen from papers [42-44], in which are reported only those activities which led to the conceptual design or even final boiler/furnace design and erection.

Our first significant result was a design of furnaces in the range of 1-5 MW$^{\text{th}}$ for the factory CER from the city of Cacak which has built more than 40 of these units on both coal and biomass (usually corn cobs) for hot air generation for drying purposes. The constructed twin FB furnaces (2×5.5 MW) in the city of Sabac are shown on fig. 8.

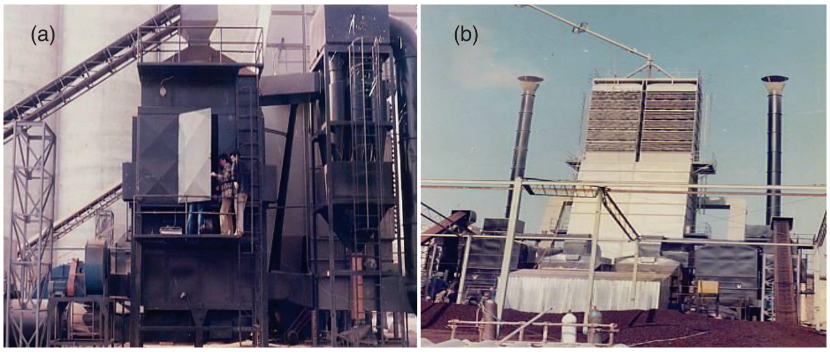

Figure 8. The fluidized bed furnace (a), and the whole facility - twin fluidized bed furnaces, vertical corn dryer with coal pile storage in front (b)

The largest boiler in whose conceptual design LTEE was involved was a 15 t/h steam boiler on wood waste in co-operation with a boiler manufacturer MINEL - Kotlogradnja from Belgrade. The unit should have been installed in a viscose fiber factory in the city of Loznica. The wood waste that was tested had a moisture content of 40-60% and the heating value in the range 6000-9000 kJ/kg. Unfortunately, due to the political and economic situation in former Yugoslavia in the early 1990's, this unit has never been built.

In co-operation with MINEL-Kotlogradnja and CER-Cacak, a 9.3 MW hot water boiler was reconstructed in LTEE [43], as a demonstrational unit and as well a 2 × 2.4 t/h steam fluidized bed boilers were erected in OTEKS factory in Ohrid, Macedonia [44]. Trial runs were performed with these twin units but they never started with regular operation, again due to the situation in former Yugoslavia in late 1980's and early 1990's. A design of 2 × 2.5 MW hot water fluidized bed boilers was also offered to the boiler house Kraljevica in the city of Zajecar.

The highest acknowledgement for our research and application achievements in the FBC field was obtaining the most important annual reward of the city of Belgrade, the so called *October reward of the city of Belgrade* in 1986 which was assigned to the LTEE research team (Lj. Jovanović, B. Arsić, D. Dakić, B. Grubor) headed by Simeon Oka, along with Milos Urosevic from CER factory and Ilija Kovacevic from MINEL-Termoremont. Prof. Simeon Oka received this same award in 1994 for his book in which main results of R&D of FBC technology up to then in LTEE was presented.

Conclusions

This paper gives an overview of the activities and obtained results in the area of R&D of FBC technology in the Laboratory for Thermal Engineering and Energy of the VINCA Institute of Nuclear Sciences, starting from mid-70's up to present days. This R&D program encompassed practically all areas needed for technology implementation: specific applied research, modeling of the overall processes associated with FBC, as well as fuel testing and involvement in specific design, erection and verification of FBC units.

Our achievements in the area of applied research such as characterization of limestones and bed agglomeration and sintering were notable in literature. In spite of the fact that former Yugoslavia was not a highly developed country and that our R&D program was only modestly funded, our results and co-operation with domestic producers enabled them to master the production of FBC furnaces/boilers, positioning Yugoslavia in early 1980's among those few highly developed countries in which R&D led to FBC technology implementation. This has established the Laboratory for Thermal Engineering and Energy of the VINCA Institute of Nuclear Sciences as one of the most important and respectable research centers in this field in South-East Europe.

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Nomenclature

C_{fix} – fixed carbon content in coal, [%] (dry and ash free basis)
$S_{\text{tot}(\tau)}$ – total SO_2 absorbed by limestone up to time, [mol SO_2]
VM_0 – volatile matter content in coal (in fig. 8 and 9), [%] (dry and ash free basis)
Vol – volatile matter content in coal (in fig. 6), [%] (dry and ash free basis)
$X(\tau)$ – degree of sulfation up to time, defined as the used up amount of both $CaCO_3$ and $MgCO_3$ in relation their initial amounts (in moles), [%]

References

[1] Arsic, B., *et al.*: Characterization of Yugoslav Limestones in a Fluidized Bed Reactor, *Proceedings*, 4th International Conference on Fluidized Bed Combustion, London, 1989, pp. I/17/1-I/17/9

[2] Arsic, B., *et al.*, Characterization of Limestones for SO_2 Adsorption in Fluidized Bed Combustion, *Proceedings*, 5th Int. FBC Conference, London, 1991, pp. 171-177

[3] Schouten, J. C, Van Den Bleek, C. M., The D.U.T - SURE Model: A Simple Approach in FBC Sulfur Retention Modeling, *Proceedings*, 9th International Conference on Fluidized Bed Combustion, 1987, pp. 749-761

[4] Lyngfelt, A., Leckner, B., Sulphur Capture in Fluidised-Bed Combustors: Temperature Dependence and Lime Conversion, *J. Inst. Energy*, *62* (1989), 450, pp. 62-72

[5] Snow, M. J. H., *et al.*, Direct Sulfation of Calcium Carbonate, *Ind. Eng. Chem. Res.*, *27* (1988), 2, pp. 268-273

[6] Grubor, B., *et al.*, Corn Cob and other Waste Biomass Combustion in FB Furnaces - Experience and Problems, *Proceedings*, 3rd European Conference on Industrial Furnaces and Boilers, Lisbon, Portugal, 1995, Vol. 2, pp.209-219

[7] Grubor, B., *et al.*, Biomass FBC Combustion - Bed Agglomeration Problems, *Proceedings*, 13th International Conference on Fluidized Bed Combustion, Orlando, Fla., USA, 1995, Vol 1, pp. 515-522

[8] Levin, E. M., *et al.*: *Phase Diagrams for Ceramists*, The American Ceramic Soc., Columbus, O. USA, 1964., Supplements 1975. and 1981

[9] Covey, G. H., Algar, V. H., UK Patent application, 6 B 2019370 A, 1978

[10] Grubor, B., *et al.*, Research And Development Of BFBC Hot-Gas Furnaces And Boilers Burning Agricultural Waste Biomass - Achievements And Problems, *Proceedings*, 39th IEA Fluidized Bed Conversion Meeting, Madrid, Spain, 1999, pp. 23-30

[11] Chandran, R. R., *et al.*, A New Method for AFBC Fuels Characterization, *Proceedings*, IX Int. ASME FBC Conference, Boston, Mass., USA, 1987, Vol. 1, pp. 292-299

[12] Chandran, R. R., *et al.*, Ranking Fuels for utility-Scale AFBC Application, *Proceedings*, X Int. ASME FBC Conference, San Francisco, Cal., USA, 1989, Vol. 1, pp. 313-322

[13] Daw, C. S., *et al.*, FBC Engineering Correlations for Estimating the Combustion Efficiency of a Range of Fuels, *Proceedings*, X Int. ASME FBC Conference, San Francisco, Cal., USA, 1989, Vol. 1, pp. 305-312

[14] Peeler, J. P. K., *et al.*, Characterization of Coals for Fluidized Bed Boilers, CSIRO, Report VI6/520, Australia, June 1990

[15] Daw, C.S., *et al.*, Characterization of Fuels for Atmospheric Fluidized Bed Combustion, *Proceedings*, XI Int. ASME FBC Conference, Montreal, Canada, 1991, Vol. 1, pp. 157-165

[16] Peeler, J. P. K., Lane, G. L., Characterization of Coals for Fluidized Bed Combustion, *Proceedings*, 12th International Conference on Fluidized Bed Combustion, San Diego, Cal., USA, 1993, Vol. pp. 649-660

[17] Preto, F., Peeler, P. K., Application of Expert Systems to Design and Control of fluidized Bed Combustors, *Proceedings*, XII Int. ASME FBC Conference, San Diego, Cal., USA, 1993, Vol. 2, pp. 769-777

[18] Oka, S., *et al.*, Experience and problems of the lignite fluidized bed combustion, *Proceedings*, 3rd. European Coal Utilization Conference, "Coal Technology Europe", Amsterdam, The Netherlands, 1983, Vol. 2, pp.87-92

[19] Oka, S., *et al.*, Problems of the Low Grade Coal Fluidized Bed Combustion, in: *Low Pollution and Efficient Combustion of Low Grade Coals* (Ed. F.Durst, K.Hanjalic and Oka, S.), Publisher Academy of Science of Bosnia and Hercegovina, Sarajevo, 1986, pp. 203-214

[20] Oka, S., Fuel Testing And Influence of Coal Characteristics on Bubbling FBC Boiler Design, Invited lecture, *Proceedings*, 1st South-East European Symposium on Fluidized Beds in Energy Production, Chemical and Process Engineering and Ecology, Ohrid, Macedonia, 1997, Vol. 2, pp. 223-242

[21] Mladenovic, M. R., *et al.*, Combustion of low grade fractions of Lubnica coal in fluidized bed, *Thermal Science*, *16* (2012), 1, pp. 295-309

[22] Milica, R. M., *et al.*, Combustion Of Hazelnut Shells In A Fluidized Bed, *Proceedings*, International Conference „Power Plants 2016", Zlatibor, Serbia, 2016, ISBN 978-86-7877-027-2

[23] Grubor, B., *et al.*, Fluidized Bed Incineration Tests Of Toxic Liquid Waste, *Proceedings on CD*, 15thInt. Conf. on Fluidized Bed Combustion, Savannah, Geo., USA, 1999, paper no. FBC99-0121

[24] Mladenovic, M. R., *et al.*,Vertical Temperature Profile in the Installation for the Combustion of Waste Fuels in the Fluidized Bed Furnace, *Proceedings on CD*, 15th Symposium on Thermal Science and Engineering of Serbia, Sokobanja, Serbia, 2011, ISBN 978-86-6055-018-9, pp. 490-499

[25] Mladenovic, M., *et al.*, Paper Waste Combustion in a Demo-Industrial Facility with FB, *Proceedings on CD,* International Conference Power Plants 2012, Zlatibor, Serbia, 2012, pp. 1220-1236., ISBN 978-86-7877-021-0

[26] Oka, S., *et al*., Investigation of The Suitability of Serbian Lignites for Burning in CFBC Boilers, *Proceedings*, FBC18: 18th International Conference on Fluidized Bed Combustion, Toronto, Canada, 2004, Paper No. 51

[27] Barisic, V., *et al*., Estimation Of Coal Pyrolysis Kinetics By Thermogravimetric And Fluidized Bed Experiments, *Proceedings*, 2nd Symposium of South-East European Countries on "Fluidized Beds in Energy Production, Chemical and Process Engineering and Ecology", Yugoslavia, pp. 1999, 63-76

[28] Barisic, V., Kinetics of Pyrolysis of Coals of Different Age, M. Sc. thesis, Chemistry Faculty, (in Serbian) University of Belgrade, Belgrade, 2000

[29] Oka, S., Fluidized Bed Combustion, ed. E. J. Anthony, Marcel Dekker, Inc., New York, Basel, 2003

[30] Mladenovic, M., Investigation of the Thermomechanical Processes During the Decomposition of High Density Liquid fuels in Fluidized Bed, (in Serbian) Ph. D. thesis, Mechanical Engineering Faculty, University of Belgrade, Belgrade, 2013

[31] Sarofim, A. F., Beer, J. M., Modeling of Fluidized Bed Combustion, *Proceedings*, 17th Symposium (International) on Combustion, Leeds, England, Vol. 17, pp. 189-204, 1979

[32] La Nause, R.D., Fundamentals of Coal Combustion in Fluidized Beds, *Chem. Eng. Res. Des*., *63* (1985), Jan., pp. 3-33

[33] Brem, G., Mathematical Modeling of Coal Conversion Processes, Ph. D. thesis, University of Twente, Enschede, Holland, 1990

[34] Van Den Bleek, C. M., *et al*., Documentation of IEA-AFBC Model Version 1.1, Report, Published inTNO Division of Technology for Society, TU Delft, The Netherlands, 1990

[35] Grubor, B., N.Verweyen: Experiences and Proposed Changes for the IEA-version 1, *Proceedings*, 17th IEA-AFBC Meeting, Amsterdam, The Netherlands, 1988, pp. 21-25

[36] Grubor, B., Combustion Tests With Yugoslav Coals - Comparison With The IEA Model, Russ. *J. Eng. Thermophys.*, *5* (1995), 4, pp. 371-385

[37] Grubor, B., Verweyen, N., Description of a New Devolatilization Scheme, *Proceedings*, IEA-AFBC Meeting, San Francisco, Cal., USA, 1989, pp. 15-19

[38] Grubor, B., Verweyen, N., Implementation of Overbed Feeding of Coal in the IEA-Model, *Proceedings*, 19th IEA-AFBC Meeting, Goteborg, Sweden, 1989, pp.42-45

[39] Grubor, B., Research and Mathematical Modeling of Processes During Combustion of Coal in Bubbling Fluidized bed, (in Serbian), Ph. D. thesis, Mechanical Engineering Faculty, University of Belgrade, Belgrade, 1992

[40] Oka, S., *et al*., The Methodology for Investigation of Fuel Suitability for FBC and Results of Comparative Study of Different Coals, *Proceedings*, Fluidized Bed Combustion In Practice: Clean, Versatile, Economic?, London, UK, 1988, pp. I/8/1-19

[41] Oka, S., *et al*., Investigation of the Suitability of Serbian Lignites Kolubara & Kovin for Burning in CFBC Boilers, *Proceedings*, 4th International Symposium of South-East European Countries (SEEC) on Fluidized Beds in Energy production, Chemical and Process Engineering and Ecology, Thessaloniki, Greece, 2003, pp. 11-21

[42] Grubor, B., *et al*., Boiler on Wood Waste in Factory "Viskoza" in Loznica, (in Serbian), *Procesna Tehnika*, (1992), 3-4, pp. 91-93

[43] Dakic, D., *et al*., Start-up and Trial Operation of a Reconstructed 9.3 MWth FBC Boiler (I Part), (in Serbian) Internal Report No. IBK-ITE-575, 1986

[44] Grubor, B., *et al*., Industrial FBC Steam Boilers with Capacity 2x2.4 t/h, (in Serbian) *Special Issue Termotehnika, 16* (1990), 3/4, pp. 285-292

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