RESEARCH IN THE AREA OF FBC COMBUSTION IN THE LABORATORY FOR THERMAL ENGINEERING AND ENERGY – PART A: ACHIEVEMENTS IN TARGETED FUNDAMENTAL RESEARCH

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The paper gives a review of the most important results of extensive targeted fundamental research program on fluidized bed combustion in the Laboratory for Thermal Engineering and Energy of the VINCA Institute of Nuclear Sciences. The paper presents a detailed overview of research activities from the beginning in the second half of the 1970' up to present days. Starting with the motives for initiating the investigations in this field, the paper highlights various phases of research and points out the main results of all research activities, not only the ones that are focused in this paper. Targeted fundamental research topics that are overviewed in this paper are heat and mass transfer, coal particle fragmentation, char particle combustion, sulfur self-retention by coal ash itself, as well as CFD modeling.

Key words*: FBC combustion, fundamental research, heat and mass transfer, char combustion, coal particle fragmentation, sulfur self-retention, CFD modeling*

1. Introduction

The technology of FBC combustion is named after the state of the matter within a boiler/furnace (bed of granular inert material suspended by a flow of air) which has a profound influence on how the process of combustion is managed and on all aspects associated with combustion such as fuel preparation, storage and feeding, methods and techniques of complying with stringent regulations on flue gas composition and ash management.

The development of this technology started in 1922 with the Winkler patent for gasification of lignite, but more than forty years passed before it was used solely for combustion, i.e. when several coal firing units were commissioned in mid-60s. The ability to burn low-grade fuels with inherently lower emissions (or, if needed, simpler and cheaper additional emission management techniques) drew attention of policy makers around the world and many countries included FBC combustion technology in their energy related R&D programs. This resulted in relatively fast diffusion of this technology which diverged in two main types: bubbling fluidized bed (BFB) and circulating fluidized bed (CFB) combustors, the basic difference being that the air velocity in CFB is higher resulting in entrainment of solids which are then separated from flue gas and returned to bed (hence the term "circulating"). R&D of FBC began in Europe but very soon spread to USA, China and elsewhere. CFB development is more focused in Europe than in USA, where the efforts on BFB still prevail. BFB technology is most suited for applications of up to several hundred MW_{th} in industry as well as for combustion of waste materials, while the CFB variant can easily achieve up to 1000 MW_{th} and more, enabling it to be used for power generation as well.

It is interesting that the initial fast diffusion of BFB boilers occurred in China, which claimed to have over 2000 BFB boilers operating in the early 1990s, but soon this technology diffused elsewhere, especially in Finland, Sweden, India, and USA. Today, the FBC technology can be considered as a mature technology for cogeneration and industrial sized applications. The potential market for BFB up to 2020, on a worldwide basis, is estimated of some 100-200 GW_{th} of additional capacity, primarily coal-fired and concentrated in China, USA and India. Much higher figures are expected for CFB in the area of coal-fired electricity generation. The demand for new capacity is estimated over 1500 GWe up to 2030. Such impressive diffusion of FBC technology is a direct consequence of huge R&D efforts mostly in highly developed countries, amply funded both by government and private sector [1].

The aim of this paper is to give an overview of specific targeted fundamental research in the area of FBC combustion in the Laboratory for Thermal Engineering and Energy of the VINCA Institute of Nuclear Sciences (in further text: LTEE). These research activities were conducted as a scientific support to our R&D of FBC technology which will be overviewed in next paper.

2. Motives and Historical Overview of FBC R&D in LTEE

The oil crises in mid-1970’ has emphasized the need to employ the following principles in many national energy policies, including in former Yugoslavia:

- ✓ Independence on import of energy or fuels. This implies: optimum fuel mix, diversity of fuels suppliers, optimal use of domestic energy resources, maximal use of renewable energy resources and of unconventional fuels such as low quality coals (high sulfur, high ash, high moisture), coal washery and separation rejects, biomass, industrial and municipal wastes.
- ✓ Development and use of clean combustion technologies. Energy technologies have to be: highly efficient (combustion efficiency >99%, boiler efficiency >85%), with high fuel flexibility (burning at the same time or consecutively different fuels), wide range of load following (1:5) and low emissions (SO_2<200ppm, NO_x<200ppm).
- ✓ Protection of environment becomes one of the main issues in energy production.
- ✓ Energy efficiency and specific energy consumption per unit production is emphasized.
- ✓ Sustainable energy development became most important including the issue of global warming.

The analysis and review of the combustion technologies used in this region as well as the emerging combustion technologies in the world led us in LTEE to the conclusion that FBC combustion technology is one of the most promising new technologies for us, taking into account the above principles as well as the fact that our coal is mostly in the form of low grade lignite. This has been emphasized by the fact that the local producers of industrial furnaces and boilers have expressed the need for new technologies that may substitute oil with domestic low quality coals, biomass and wastes.

Our concept of FBC R&D program was as follows [2]:

- ✓ **in the first phase of R&D program** to organize applied research and to develop engineering calculation methods,

- ✓ to investigate processes most important for burning domestic coals (lignites and high volatile bituminous coals),
- ✓ for calculation of other processes in the furnace data from literature can be used,
- ✓ to design and build experimental facilities for applied research and technology development - experimental and pilot furnaces,
- ✓ to experimentally determine the necessary data for the choice of concept, design and calculation of bubbling FBC boilers and hot-gas generators,
- ✓ verification of our laboratory tests, calculation and scale-up methods by testing of demonstration units,
- ✓ **in the second phase of R&D program,** which can start before the end of the 1st one, to start with fundamental research on small experimental facilities, and then
- ✓ develop mathematical models for prediction of process parameters in FBC furnaces, as a basis for engineering calculation and optimization.

In the late 1970' a FBC research team of five full time researchers has been established in LTEE headed by Prof. Simeon Oka [2]. An intensive R&D program has been established starting with literature survey, building up our experimental basis ranging from small units for hydrodynamic studies at ambient conditions and ending up in 1980 with a 200 kW_{th} furnace enabling steady state combustion tests. As stated before, the main aim of this R&D phase was to help the domestic industry to master this technology and to build their capacity to design and produce FBC furnaces. This goal was fulfilled in 1981 when CER factory from Cacak erected 2x5.5 MW furnaces burning coal to produce hot air for drying purposes.

2.1. International Cooperation

From the very start, it was clear that we had to rely on FBC technology research and development results in the developed countries of the world. Therefore, our policy during all of the period 1976 – 2000 was to closely cooperate with research teams from abroad, participate in international projects if possible, and send our younger staff to foreign universities and institutes for training purposes. It goes without saying that this also includes an extensive participation on international conferences and seminars. We had a very good and intensive cooperation with the following universities and institutes:

- Chalmers university of Technology, Sweden
- University in Aachen, Germany
- Institut-teplomosso obmena, Minsk, Belarus
- Abo University, Turku, Finland
- Institut teplofiziky, Novosibirsk, Russia

In these institutions some of our staff has visited and stayed from several months up to one year and participated in their FBC investigations.

By far the most important international cooperation was our involvement in the IEA FBC Implementing agreement of the OECD countries (for cooperation in the ***field of Atmospheric Fluidized Bed Combustion in Industrial or District Heating Boilers***). It started from 1983 when former Yugoslavia gained a status as an observer but very soon gained full membership in 1986. Institute Vinca (i.e. LTEE) was chosen as the representative of Yugoslavia in this Agreement, because of its results in the field of FBC R&D, and Prof. Simeon Oka was appointed as our delegate in the Executive Committee of the Agreement (Dr. Borislav Grubor as his deputy).

Representatives from LTEE have regularly attended all Technical Meetings (twice yearly) and participated in common research activities (such as modeling of FBC processes). In the period 1990/1991 Yugoslavia coordinated the activities of this Agreement and Prof. Simeon Oka was the chairman of the Executive Committee. Even when international sanctions were introduced, the members of the Agreement allowed our representatives to attend the technical meetings, but without the right to vote. Unfortunately, neither Yugoslavia nor todays Serbia have renewed their membership in this Agreement, due to the changed political circumstances. LTEE had significant benefits from the involvement in this Agreement: our staff had the opportunity to gain insight in new developments in the field of FBC R&D and contacts were established with most prominent institutions in this field of research (which enabled our younger staff to visit them) as well as with most significant producers of the FBC technology.

In an attempt to encourage the cooperation of the countries in this region in the FBC field, LTEE was a promoter of forming an ***Agreement For Co-Operation In The Field Of Fluidized Bed Conversion Applied To Efficient And Clean Energy Production And Chemical Engineering*** of the South-East European Countries, which was signed in 1998 by 13 scientific and academic institutions from Bulgaria, Rumania, Macedonia, Greece, Bosnia and Hercegovina and Serbia. Later in 2003 the name of the Agreement was changed to ***Agreement For Co-Operation With The Aim To Research, Develop And Implement New Energy Efficient And Ecologically Acceptable Technologies Applied To Energy Production, Chemical And Environmental Engineering***. In the period 1997-2005 five symposiums have been organized under this Agreement. After 2005 the interest for this Agreement ended since many of these countries joined EU and other West Balkan countries could join the FP 5 and FP 6 projects financed by the EU Commission.

2.2. Research Phases

The first half of the 1980’ was a period when LTEE has tested dozens of fuels which enabled a formation of an extensive experimental data bank. This assisted formulation and subsequent utilization of own methodology for further fuel testing. In mid-1980’ the Institute has even started some initial R&D of CFB technology by building a cold rig for hydrodynamic tests. Unfortunately, the late 1980’ was a period of political tensions resulting in violent collapse of Yugoslavia. The industrial activity ceased and financing of our FBC R&D from industry subsided.

We seized the opportunity to step-up our fundamental research of various processes associated with FBC combustion, since practically all of the available research capacity could focus in that area, and the period of 1990’ was the most productive period in LTEE. The need for our own fundamental research using our low quality coals has been emphasized by the fact that most literature data was associated with hard coals. The noticed problems in our design and operation of early units, and the need to rectify them, only intensified the need for our better understanding of these processes and focused our research. Our research was mainly supported through projects which were financed by our Ministry and the research group in LTEE actually increased in this time period.

When the political situation in Serbia and in the region stabilized, the conditions and circumstances for R&D in the area of FBC have drastically changed. Firstly, there were practically no significant boiler manufacturers in Serbia that could support any R&D, let alone in the area of FBC.

Secondly, the energy situation in the world has also drastically changed, i.e. the focus has altered from fossil fuels to renewable sources and practically all research related to coal usage has subsided since the state policies in developed countries favored gas.

This inevitably influenced the change in program orientation of the research group in LTEE, which was up to then focused on coal utilization. Our new research policy was now oriented towards biomass and wastes, and the focus gradually switched from technology development to technology implementation. This was mainly done through participation in internationally funded projects, like FP5, FP6, and FP7 programs, financed by EU Commission, in which LTEE participated in case studies, distributive power production analysis, utilization of biomass and wastes. Our participation was in many cases based on implementation of FBC technology involving our conceptual designs and in some cases our specific technical solutions, especially in cases when agricultural biomass wastes were considered.

3. Heat and Mass Transfer

Initial heat and mass investigations in LTEE were focused on heat transfer between an immersed tube bundle and fluidized bed with the aim to be able to determine heat transfer coefficients for design purposes. At that time (early 1980') there was not enough reliable data in literature on this topic, especially for larger particles of inert bed materials and our research activities were based on the results of only several authors [3,4].

For this purpose a simple test rig was designed and built. The fluidized bed operated at near ambient temperatures. In the bed a tube bundle is submerged among which only one tube was used for measurements while the other tubes served simply to simulate the appropriate hydrodynamic conditions. By measuring the heat supplied to the cooper tube, along with the cooper tube surface temperature and the bed temperature, at steady state conditions, it was possible to determine the overall heat transfer coefficient. The experiments were done for several particle sizes of sand as inert material (in the range 0.5 – 1.5 mm), and varying the bed height, the tube bundle alignment as well as pitch ratio [5-7]. Two MSc Thesis resulted from these investigations [5, 7] which enabled us to choose the most appropriate empirical expression found in literature.

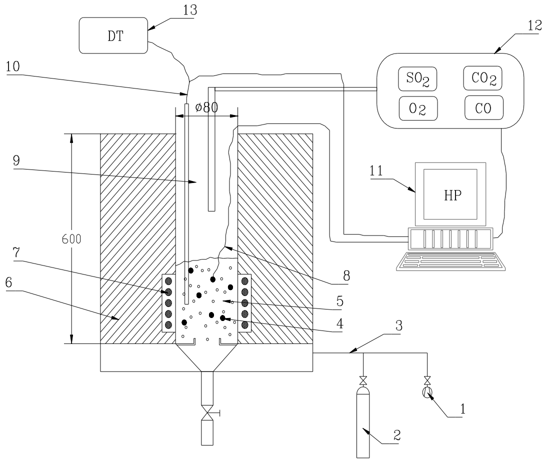
The experiments were also used to study the general hydrodynamic behavior of the bed with and without the immersed tube bundle, such as the inception of fluidization and the dilatation of the bed layer [8]. An attempt was made to study also the bubble frequency at certain locations of the bed using the light emitting and light detecting diodes. These diodes were immersed in the bed separated one from another a few millimeters. The variation in the quantity of light detected was a direct consequence of bubbles moving over these diodes.

Our later investigations shifted towards the area of heat and mass transfer between immersed particles and fluidized bed as a result of our need to improve our modeling capabilities in regard to char particle combustion. Another reason was related to our design aspirations. Namely, these investigations would enable us to choose the operating bed temperature with more confidence in cases when the fuel has agglomeration and sintering tendency. In these cases it was important that fuel particles do not surpass certain temperature limits.

The heat and mass transfer experiments were done in a small, electrically heated fluidized bed furnace under the following conditions, Fig. 1:

- mean sand (bed) particle diameter, d_p = 250, 500 and 900 μm,

- sphere diameter, d_s = 5, 10, 15, 20 mm,
- bed temperature, t_b = 300, 500, 600, 750 ^oC,
- fluidization velocity, v_f = 0.092 - 0.855 m/s.



1. Fan,
2. N_2,
3. Tubes,
4. Coal particle,
5. Fluidized bed,
6. Thermal insulation,
7. Electrical heater,
8. Thermocouple in coal particle centre,
9. Fluidized bed reactor,
10. Thermocouple in fluidized bed,
11. Data acquisition system,
12. Gas analyzer,
13. Digital thermometer.

Figure 1. Experimental fluidized bed reactor, i.d. 80 mm.

Active particle temperature was measured using 0.5 or 1 mm Cr-Ni thermocouples imbedded in the center. Air and nitrogen were used as fluidization gases. The heat transfer coefficient was determined by measuring the center of the sphere temperature variation during its heating up in the bed. This area of research was investigated by many authors in the world so we had good guidelines for our investigations [9 – 13].

On the basis of numerous experiments, the influence of particle diameter, mean size of bed particles, bed temperature and fluidization velocity on the heat and mass transfer coefficients was analyzed. Correlations for both heat and mass transfer were obtained encompassing the experimental results with an accuracy of ± 15% in the case of heat transfer, Fig. 2, and an accuracy of ± 30% in the case of mass transfer, Fig. 3 [14 - 16].

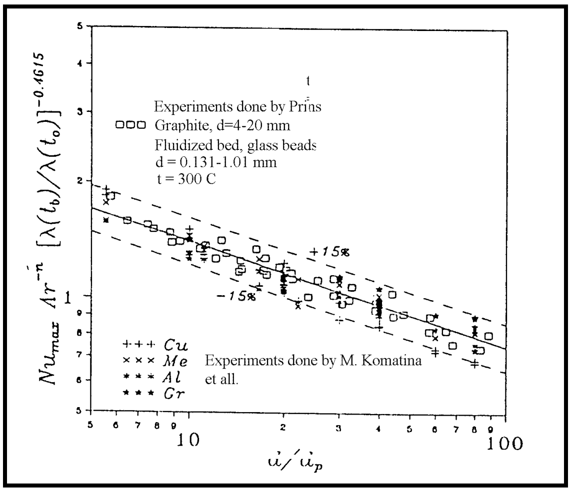


Figure 2. Heat transfer coefficient between coal particle and fluidized bed.

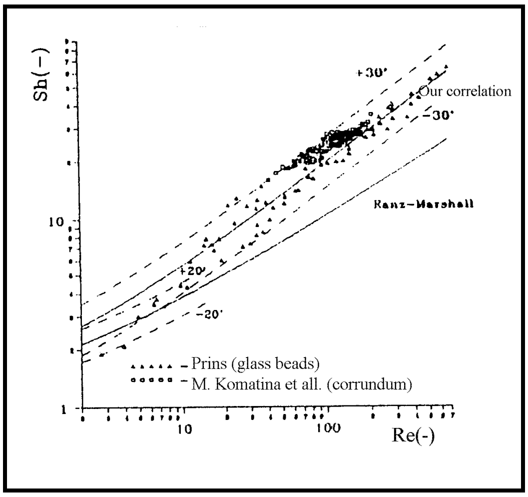


Figure 3. Mass transfer between coal particles and fluidized bed.

The experiments encompassed the coal particle heating-up and pyrolysis [16], devolatilization [17], and char combustion [11], as well as other immersed particles [19, 20]. These investigations resulted in a Ph. D. Thesis by Prof. Mirko Komatina [21] and I. Mijakovski [22], both being from other institutions but whose research in this area was closely associated with LTEE.

4. Coal Particle Fragmentation During Devolatilzation

In the very beginning of our experimental investigation of FBC single coal particle combustion we evidenced a significant degree of coal particle fragmentation after devolatilization. It was especially noticeable in our batch experiments where we could do a reliable mass balance of the batch particles before and after the process of combustion, and thus notice the change in number and size of particles.

Since we did not notice any significant attention in literature for these phenomena, apart from the work in [23], we commenced a comprehensive experimental investigation of this process in a small FB furnace, Fig. 4. Experimental procedure involved introduction of a well-defined batch of particle of a certain coal in an electrically preheated FB and flushing the whole inventory of the FB after devolatilization down in a flask quenching any further reactions. Investigation of the process of fragmentation was done by analyzing the photographs of coal particles before and after devolatilization (example shown also on Fig. 4).

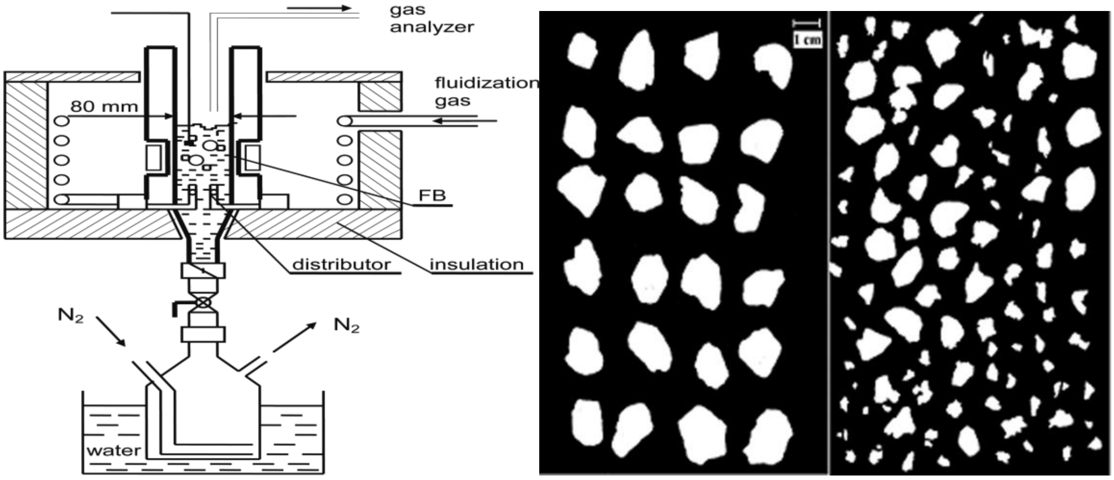


Figure 4. Experimental setup & typical photograph of coal samples before/after devolatilization.

A wide-ranging experimental data were obtained [24-27], which resulted in one MSc Thesis [25] and a Ph. D. Thesis [26]. Most importantly, it enabled us to suggest a critical fragmentation diameter, d_{cr}, as a function of the so called Pore Resistance Number, PRN, equal to the ratio of the volatile matter content and the equilibrium moisture content in coal, above which fragmentation occurs, Fig. 5 [28]. For lignites critical diameter is 10-15mm, while high rank coals already fragment when particle size is 5mm. In spite of this, fragmentation is more important for modeling of lignite combustion, because lignites are fed with particle size up to the 50 mm. High rank coals, because of low reactivity, are grinded before feeding usually to the size smaller than critical fragmentation diameter.

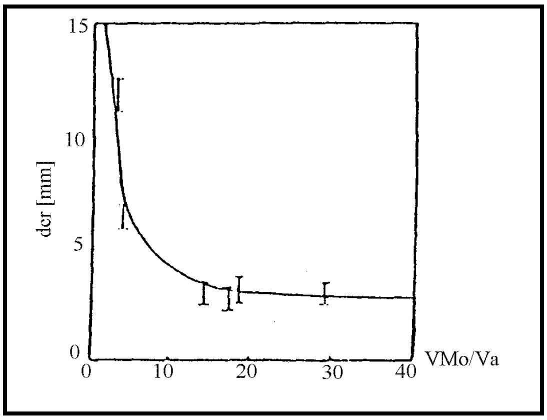


Figure 5. Critical fragmentation particle diameter for different coals [28].

Parallel to the experimental work modelling efforts were made. Initial models were of a statistical and mechanistic nature relying on the parameter d_{cr} and geometrical considerations of coal particles [28-30]. Later refinements of the model [31, 32] are based on defining two patterns of fragmentation: smaller fragments (relative to the original coal particle size), which originate from the outer shell of the particles, and larger fragments which are formed due to the fracture of the inner core. The smaller fragments are formed shortly after the coal particle has been introduced to the hot environment and are a result of thermal shock, whereas the larger fragments are formed later during devolatilization, due to the pressure build-up inside the particle.

The model input data are the coal type characteristics (volatile and carbon matter content, porosity, mean pore diameter), bed temperature, and the initial coal particle size distribution. The model output data are the resulting char particle size distribution (number and mass fractions of char particles) and the primary fragmentation parameters which are as follows [31]:

- Primary fragmentation ratio, N_f, which is defined as a ratio of the number of original coal particles and the resulting number of char particles, after fragmentation. Thus, this parameter increases with the intensity of fragmentation..
- Changing ratio of coal particle size: $Fd = \Sigma x_i d_i/d_c$. Here, x_i is the mass fraction of particles with size i, d_i is the average diameter of coal particles with size i after fragmentation, and d_c is the diameter of the original coal particles. This parameter decreases as the intensity of fragmentation increases, but may also be > 1 for coals which experience swelling during devolatilization.

- Primary fragmentation index, which is defined as a ratio of the two previous parameters. This is the most comprehensive parameter of the process, which takes into account the changes both in number and size of particles.

A procedure is defined which enables initialization, propagation, and merging of cracks. Comparison with experimental results has shown (an example given on Fig. 6) that the model gives reasonable predictions of the fragmentation process.

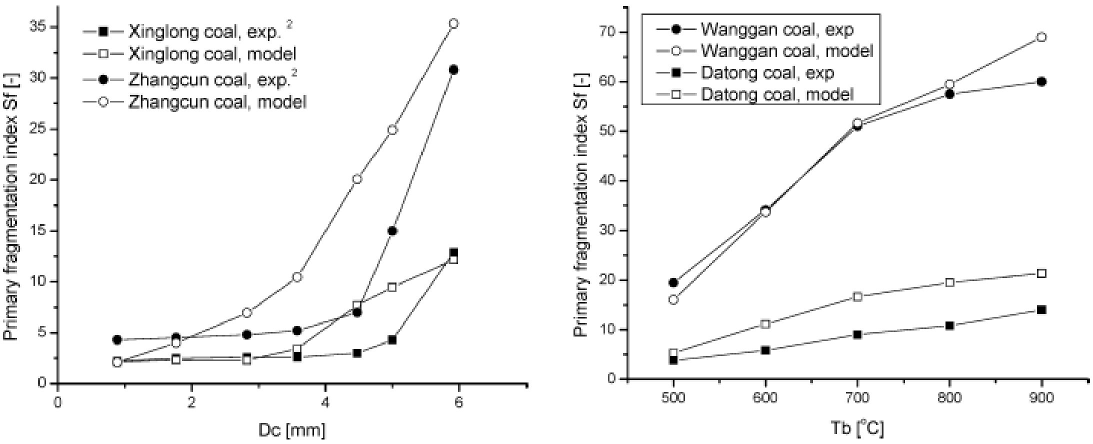


Figure 6. Primary fragmentation index, comparison with experimental results, influence of size of coal (left), influence of FB temperature (right).

Investigations in this area are continuing in the direction of determining the pressure build-up inside the coal particle during devolatilization [33] with the aim to develop an even more comprehensive model of coal particle fragmentation in the future.

5. Char Combustion

Char combustion is the most important process for the behavior and operation of the BFBC boilers, and for mathematical modeling of the processes taking place in the furnace. The main part of heat generated comes from char combustion and char residence time in the fluidized bed is significantly longer than the residence time of volatiles. Char particle size distribution and char hold up in the bed influence many other processes (attrition, elutriation of unburned particles, NO_x reduction, rate of load change, etc.). Having also in mind that coals in our region are of low grade and have certain specific characteristics which were not sufficiently investigated elsewhere, we started our own investigations. Our investigations and modeling work was based on the achievements of many authors who were dealing in this area of research at that time, pointing out the most significant for us [34-37].

Experimental investigation using batch experiments were done in a furnace with electrically heated fluidized bed, Fig. 1 [38-40]. Bed temperature and composition of combustion products were measured in time, as well as particle mass defect, particle temperature and particle porosity during combustion.

After forming a sufficient data base on char particle combustion, investigations in LTEE were broadened by including modeling of this process [41-43]. The mathematical model developed describes the dynamic behavior of porous char particle and includes only two chemical reactions: a heterogeneous reaction on the external and internal surface of the particle (Reaction 1: $C + O_2 \rightarrow x \cdot CO$

+ y·CO_2) and a homogeneous reaction within the pore volume of the particle (Reaction 2: $CO + O \rightarrow CO_2$). The particle is composed of one solid reactant (carbon), mineral matter and voidage. The environment surrounding the char particle consists of O_2, CO_2, CO, H_2O and N_2. The role of H_2O is only as a catalyst for the homogeneous reaction.

A microscopic approach to the char combustion model was adopted. Temporal and spatial evolution of char particle physical properties, heat and mass transfer characteristics, as well as temperature and gas concentrations are defined. The pore structure locally evolves proportional to the local particle conversion and affects the heat and mass transfer properties of the particle. Char particle shrinkage is allowed, the criteria being that the local degree of conversion of the external surface layer is over 98%. The temperature and gas concentration field within the char particle are defined by well-known differential equations of heat and mass conservation.

The numerical method of control volume was applied. The char particle is divided in numerous spherical shells. The change of physical properties of the solid material, the heat and mass transfer characteristics, and temperature and gas concentrations are defined for each spherical shell.

Testing of the model showed that it can closely predict the combustion of many tested coals, both the temporal changes of gaseous combustion products as well as the over-all burn-out times provided that there was no fragmentation of char particle during combustion. Since good predictions were obtained in the range of 500 – 700 ^{o}C FB temperatures, it implies that all three main mechanisms, namely mass and heat transfer and reaction kinetics, are well balanced in the model, resulting in a good assessment of the degree of either kinetic or internal diffusion control for any set of specific combustion conditions and a wide variety of coal char characteristics. Some typical results of these comparisons are shown on Fig. 7 [42, 43]. One MSc Thesis [38] and one Ph. D. Thesis [41] resulted from research in the area of coal and char combustion.

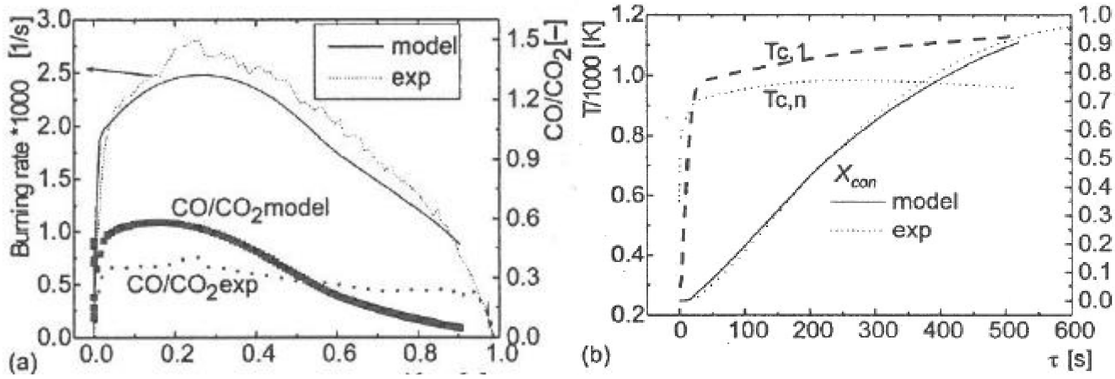


Figure 7. Experimental results and model predictions for 5 mm brown coal Aleksinac at 600 ^{0}C, (a) Burning rate and CO/CO_2 ratio, (b) Char particle temperature and carbon conversion degree versus time.

6. Sulfur Self-Retention by Coal Ash

The initiation of investigation of the process of sulfur self-retention (SSR) by coal ash itself came as a result of inability to explain seemingly erroneous experimental results related to SO_2 emissions during steady-state combustion tests with various domestic and foreign coals. Literature survey has shown that other authors also noted the SSR phenomena, in some cases even up to 60% in bubbling fluidized beds and up to 90% in CFB conditions, but it was not possible to correlate or explain the level of our obtained emissions with the sulfur content in coals nor with any other coal characteristic. We could not use directly the results of investigations of this process by other authors [44-49] due to significant differences in characteristics of the investigated and our coals. This research topic in LTEE was initiated Dr Borislav Grubor but most of the results were obtained through the work of Dr Vasilije Manovic whose both masters [50] and Ph. D. Thesis [51] are focused on this topic.

The initial research program was focused to the study of the chemistry of sulfur, namely the transformation of sulfur forms during the various processes that coal particles undergo during devolatilization and char combustion. These investigations were initially done by placing samples of various coals, in narrow sieve class ranges, in a laboratory oven which was preheated to temperatures in the range 750 – 900 °C. Sulfur forms in the coal and sulfur in the resulting ashes were determined.

The total Ca content in ash of coals, in all sieve classes, was determined using standard atomic absorption spectroscopy (AAS), but only the Ca that can be extracted by hot HCl solution is considered as active Ca.

Numerous experiments and studies have shown that the released amount of sulfur during devolatilization may be determined by taking into account the following assumptions (approximations) in regard to the transformation of various sulfur forms during devolatilization:

1. The total amount of sulfate sulfur in coal remains in char.
2. One half of the pyritic sulfur (S_p) is released during devolatilization as a result of the decomposition of pyrite according to the following reaction: $FeS_2 \rightarrow FeS + S$.
3. The amount of organic sulfur released with the volatiles is proportional to the organic sulfur content in coal (S_o). The proportionality coefficient may be defined as equal to $VM/(C_{fix}+VM)$, implying that the organic sulfur is proportionally distributed.

Based on the previous assumptions, the following correlation was proposed for evaluating the total sulfur that is released during devolatilization:

$$S_{R,VM} = 0.5 \cdot S_P + \frac{VM}{C_{fix} + VM} \cdot S_O$$

Our research results in this area, including the verification of the above correlation, have been reported on many occasions [52-55]. On Fig. 8 the comparison is given between experimentally obtained data for six coals (from Serbia and former Yugoslavia) as well as the data found in the literature on the amount of sulfur released during devolatilization and the calculated values according to the proposed correlation. Regardless of the above simplifying assumptions, it may be seen that there is a quite good agreement between the calculated and experimentally obtained values.

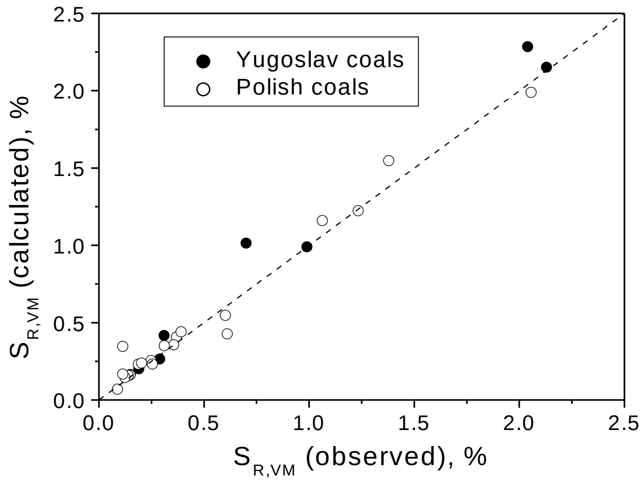


Figure 8. Experimental and calculated amount of released sulfur during devolatilization (%,dry coal basis), for Yugoslav (•) and Polish (O) coals, [55].

Taking into account the sulfur retention efficiency during char combustion (η), the amount of sulfur released during the whole process of combustion is equal to:

$$S_{R,T} = (1-0.5\eta)\cdot S_P + \left[1-\frac{C_{fix}}{C_{fix}+VM}\cdot\eta\right]\cdot S_O + (1-\eta)\cdot S_S$$

In order to evaluate the overall sulfur retention in ash during the whole combustion process it remains to determine the sulfur retention efficiency during char combustion η, stated in the above equation. Further laborious studies showed that this could not be accomplished by deriving some simple empirical correlation as in the case of devolatilization process, but that the complexity of the process requires modelling approach.

The developed model for SSR is coupled with the previously described model for char combustion [42, 43]. The main role of the char combustion model is to generate time and special data on carbon conversion since the rate of SO_2 formation is proportional to the rate of carbon conversion.

It is assumed that the active part of CaO in coal, formed as a result of $CaCO_3$ decomposition and combustion of Ca-containing organic compounds, is the only base oxide that contributes to sulfur retention. Our procedure for evaluation of the active part of CaO, as well as a more detailed discussion about Ca forms present in coal, its distribution and activity has been presented on several occasions [53-55].

The reaction of SO_2 with CaO (sulfation) is first order with respect to SO_2 and zero order with respect to O_2. The sulfation rate is taken into account in a simplified manner by assuming that the sulfation rate decreases proportionally with the conversion degree of CaO.

A novel approach has been applied for modeling of SSR [56-59], closely related to the grain model used for SO_2 retention by limestone addition. It is assumed that after devolatilization all of the active Ca is present in the form of CaO grains of the same initial radius ($R_{G,0}$), which are uniformly distributed throughout the char volume. An unreacted shrinking core model is adopted for the following reaction between the CaO grains and SO_2:

$$CaO + SO_2 + \tfrac{1}{2}O_2 \rightarrow CaSO_4$$

The model also takes into account thermal decomposition of $CaSO_4$ as well as reduction of $CaSO_4$ by carbon monoxide with appropriate correlations of the Arrhenius type. Temperature and concentration profiles along the char particle radius are obtained by solving the appropriate differential equations for heat and mass balance.

The results of batch combustion tests in both a fluidized bed (FB) reactor and in a laboratory oven for two Serbian coals, Kolubara and Bogovina, were used for experimental verification of the model. From Fig. 9a it can be seen that a substantial part of sulfur evolves during devolatilization and that the concentrations of SO_2, monitored at top of the bed, are significantly higher than during char combustion. Since the evolution of SO_2 during devolatilization is taken into account in the model by the above given correlation, the comparison between experimental and model predicted SO_2 concentrations was done only during the char combustion, Fig. 9b. It may be seen that the model can adequately predict the evolution of SO_2 during char combustion.

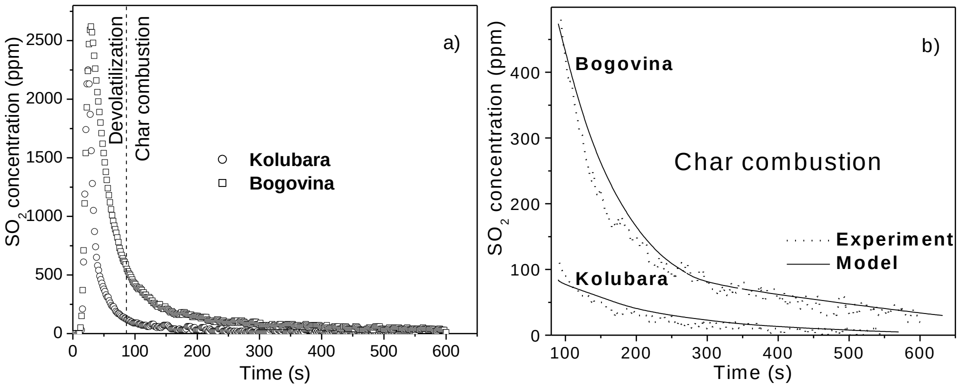


Figure 9. SO_2 concentration at top of the bed during batch combustion of coal, T_{bed} = 750 ^{o}C R_{Ch} = 2 - 3.5 mm: a) experimental values during the whole process of combustion, b) experimental values and model predictions during char combustion [56].

7. CFD modeling

In parallel to modeling overall processes during steady-state FBC combustion of coal, a more detailed modeling of a gas-particle system has been undertaken, with the aim to broaden our modeling capabilities to liquid and gas FBC combustion and thermal disintegration (incineration) of industrial wastes and by-products. For these purposes two types of CFD fluidized bed models were developed: CFD fluid-porous medium model, developed for stationary 2D predictions of fluidized bed and CFD two-fluid Euler-Euler unsteady granular model of a fluidizing furnace. As guidelines we used the investigations of other prominent authors in this area [60-63].

For the fluid-porous medium model of the fluidized bed, the dense phase is considered as a fixed porous medium, while gas-particle interactions and bubbling phase are modeled regarding balance of friction forces between gas and particles, including the effects of the FB particle collisions [64, 65].

The two-fluid Euler-Euler model of a fluidizing furnace implies unsteady modeling of the gas and particle velocities in two-phase granular flows based on analogy with kinetic theory of gases (KTGF), incorporating energy equations of the gas and particle phase, as well as transport equations of chemical components with the source terms due to the conversion of components [66-68].

Chosen was the Euler-Euler fluidized bed modeling approach which considers the gas and FB dense phase (gas-particle system under conditions of the minimum fluidization) as two fluids with different characteristics. In the transport equations for transfer of momentum of the effective fluid (the FB dense phase), fluid-particle interactions in conditions of the minimum fluidization velocity are modeled, as well as the interaction between the particles themselves. In the Eulerian-Eulerian approach all phases have the same pressure and that is the pressure of the continuous - primary phase. This model solves the continuity and momentum equations for each phase, and tracks the volume fraction. The additional transport equation for the granular temperature (which represents the solids fluctuating energy) is solved and the solids bulk and shear viscosity are determined using the kinetic theory of gases.

For modeling the interactions between gas and particle phases, within the suggested Euler-Euler granular approach to fluidized bed modeling, the routines incorporated in the modules of the commercial CFD software package FLUENT 6.3.26 were used. This code allows presence of several phases within one control volume of the numerical grid, by introducing the volume fraction of each phase. The solid phase represents a granular layer made of spherical particles, with uniform diameters. The mass and momentum conservation equations are solved for each phase separately. The comprehensive model of the complex processes in fluidized combustion chamber incorporates, besides gas and particular phase velocity fields' prediction, also the energy equations for gas and solid phase and the transport equations of chemical species conservation with the source terms due to the conversion of chemical components. A more detailed explanation of our modeling approach is given in several publications [64, 66, 69].

This model has been applied to combustion in the freeboard [69], gaseous fuel jet injection into a FB furnace [64, 65], and combustion of liquid fuels in FB [66-68]. The model has been verified comparing it with experimental results, including our own as well as data from literature. On Fig. 10 shown are the results of gas temperature distributions during combustion tests with diesel fuel and on Fig. 11 the comparison between calculated and experimentally obtained temperature profiles along the FB height [67].

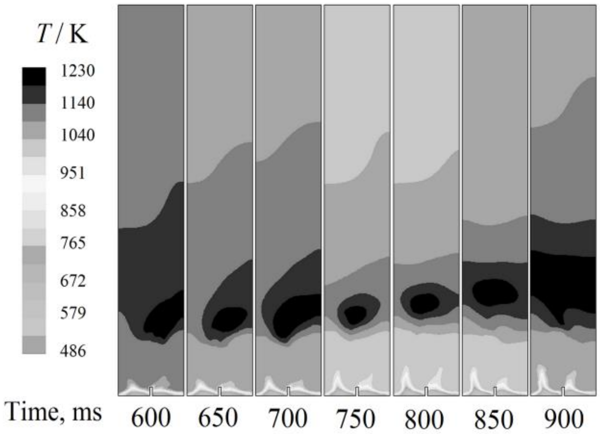


Figure 10. Calculated gas temperature distributions during combustion of diesel fuel.

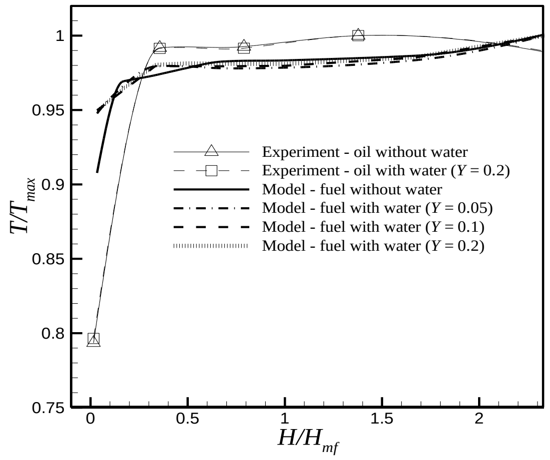


Figure 11. Normalized temperature profiles along fluidized combustor height.

8. Conclusions

This paper gives an overview of targeted fundamental research program in the area of FBC combustion in the Laboratory for Thermal Engineering and Energy of the VINCA Institute of Nuclear Sciences, starting from mid-70s up to present days. These research activities were conducted as a scientific support to our R&D of FBC technology with the aim to assist the domestic industry in acquiring this technology and implementing it in former Yugoslavia.

The targeted fundamental research in areas of heat and mass transfer, coal particle fragmentation, char particle combustion, sulfur self-retention by coal ash itself, and CFD modeling, coupled with extensive experimental work, resulted in notable amount of published data, developed correlations as well as in models of these processes with relatively good ability to predict these processes in real conditions. More than 15 researchers were engaged in these activities resulting in 5 MSc and 6 Ph. D. Thesis and one book.

R&D activities and obtained results in the area of FBC combustion have 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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10. Nomenclature

Ar - Archimedes number ($= gd_p^3\rho_p/\rho_g\nu_g^2$), [-]
C_{fix} - fixed carbon content in coal, [%] (dry coal basis)
d - diameter of immersed particle, [m]
d_{cr} - critical fragmentation particle diameter (above which fragmentation occurs), [mm]
d_p - mean bed particle diameter, [m]
D - diffusivity of naphthalene vapor, [m2s]
d_i - diameter of coal particle of size class “i” after fragmentation, [mm]
d_c - diameter of original coal particle, prior to fragmentation, [mm]

F_d - changing ratio of coal particle size (= $\Sigma x_i d_i/d_c$), [-]
h_{max} - maximum coefficient of heat transfer to coal particle, [Wm-2K-1
N_f - primary fragmentation ratio (= N_{out}/N_{in}), [-]
N_{in} - number of coal particles of size class ”i”, [-]
N_{out} - number of char particles after primary fragmentation of coal particle size class ”i”, [-]
Nu_{max} - maximum Nusselt number based on bed particle diameter (= $h_{max} d_p/\lambda_g$), [-]
PRN - pore resistance number (= VM_0/V_a), [-]
Re - Reynolds number (= $v_f d_p/\nu_g$), [-]
S_f - primary fragmentation index (= N_f/F_d), [-]
Sh - Sherwood number (= kd/D), [-]
S_O - organic sulfur content in coal, [%] (dry coal basis)
$S_{R,T}$ - released sulfur during coal combustion, [%] (dry coal basis)
$S_{R,VM}$ - released sulfur during devolatilization, [%] (dry coal basis)
S_S - sulfate sulfur content in coal, [%] (dry coal basis)
S_P - pyritic sulfur content in coal, [%] (dry coal basis)
t_b - fluidized bed temperature, [^{o}C]
t_o - ambient temperature, [^{o}C]
v_f - fluidization velocity, [ms-1]
V_a - equilibrium moisture content in coal, [%]
VM - volatile matter content in coal, [%] (dry coal basis)
VM_0 - volatile matter content in coal, [%]
x_i - mass fraction of coal particles after fragmentation with size class “i”, [-]

Greek symbols
ρ_p - particle density of bed material, [kgm-3]
ρ_g - density of fluidizing gas, [kgm-3]
λ_g - thermal conductivity of fluidizing gas, [Wm-1K-1]
ν_g - kinematic viscosity of fluidizing gas, [m2s]
η - sulfur retention efficiency during char combustion, [-]

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