Procedures for recovering the residual coal from bottom ash

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***Abstract*—** The carbon content in ash differs depending on the quality of the coal, the efficiency of the combustion plant, the combustion parameters and so on. The separation of unburned coal from ash can be done for various purposes - to reduce the unburned coal content before its use (eg for cement production), to collect uncharged coal to further study its characteristics or to prepare some absorbents, graphite materials, etc. The choice of the separation procedure is therefore given by the purpose of the separation, the properties required for unburned carbon, or by the subsequent intended use. The UCB research team analyzed twelve pre-concentration procedures for residual carbon from bottom ash. The procedures and the methodology used for comparison are pointed out.

***Keywords*—** bottom ash, char, pre-concentration procedures.

I. Introduction

According to the International Energy Agency and the World Coal Association, coal provides about 30% of the global primary energy needs. The total coal production in 2012 reached 7830 Mt and it has been estimated that current coal reserves are sufficient to meet at least 100 years of supply at this level of production [1].

The carbon content in ash differs depending on the quality of the coal, the efficiency of the combustion plant etc. Considering the most favorable case, where the average carbon residues content in ash would be 1%, it would result in about 8 million tons of carbon residues per year. Since the carbon content in ash is generally higher (often 3-5%), and given that the use of modern low-NOx burners rather complicates the effort to decrease it, the annual carbon residues in ash is estimated to be of tens of millions of tons [1].

In addition to the priority aspect of energy loss due to incomplete coal combustion, there are two main areas of research to be given special attention today:

- The higher level of carbon content in ash prevents the subsequent use of these ashes as an addition in Portland cements and in building materials industry, also involving increased cost of waste transporting and / or storing. From this point of view, the carbon residues from the ash are an unwanted component whose percentage should be reduced by an optimized combustion process or by efficient separation techniques.
- Because of the promising characteristics of unburned carbon, there is a strong tendency to find a feasible use of this material, for example, for the preparation of efficient absorbents, graphite substitutes, and so on.

II. Problem Formulation

The separation of unburned coal from ash can be done for various purposes - to reduce the unburned coal content before its use (eg for cement production), to collect uncharged coal to further study its characteristics or to prepare some absorbents, graphite materials, etc. The choice of the separation procedure is therefore given by the purpose of the separation, the properties required for unburned carbon, or by the subsequent intended use [2].

The coal ash storage raises concerns in terms of economy and environment (e.g. a 330 MW pulverized fuel unit consumes around 1,000 ton/h of coal and generates ca. 400 tons of ash and slag at the same time). It is true that a part of fly ash that may be reintroduced into the economic circuit by using it in cement industry. However, only 5-8% of the coal ash generated is used currently in Romania, the rest being stored in large landfills, usually built through major changes of the natural landscape and containing tens or even hundreds of millions tones of ash [3]. Therefore, several actions are being taken in Romania in order to decrease the amount of ash landfilled, and to raise awareness concerning the need to revise legal provisions (e.g. actually the use of ash in road construction is still banned, without distinguishing between fly ash (chemically active ash that can generate important secondary phenomena by hydration processes) and bottom ash (chemically inert and with physical-chemical properties very close to those of natural granular aggregates, like sand) [4,8].

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The University "Constantin Brancusi“ of Targu Jiu (UCB) is actively involved since 2011 in an research effort to detect efficient directions to recover the coal ash generated by thermal power plants, prevalent in the Oltenia industrial area, particularly for using them as alternative raw material in the building industry [5]. Therefore, UCB continue and widens the efforts to recover coal ash through its participation in the European Project CHARPHITE consortium under the scope of the “Third ERA-MIN Joint Call (2015) on Sustainable Supply of Raw Materials in Europe.

The main goal of the project is to use the carbonaceous solid residue (char) from Oltenia bottom ash as substitution material for natural graphite in cutting-edge energy technologies, such as catalysts for electrochemical reactions in cell batteries or hydrogen and oxygen production by water electrolysis [6].

The UCB’s research team contribution in this project mainly aims the separation of the char from fresh and landfilled bottom ash, and further assessment and utilization of the “char-free” coal ash.

III. Methodology and Procedures

Generally, the carbon residues (char) fractions can be separated from other fractions using wet processes (gravimetric separation, froth flotation and hydrophobic agglomeration in the upper hydrocarbon medium) or dry techniques (sieving, incipient fluidization and tribo - electrostatic separation). Compared to wet separation procedures, dry separation techniques do not pose a risk of leaching of soluble or contamination specimens, which may be problematic if, for example, the chemical composition is studied.

Under the scope of CHARPHITE project, we initially collected 100 kg of fresh bottom ash from the coal fired boilers Govora Combined Heat and Power Plant [7].

Once collected, the ash samples were immediately closed in plastic boxes, to preserve their original properties until the laboratory testing to determine the moisture concentration, bulk density and granulometry. Moisture was determined by drying the sample in an electric oven (150 liters capacity) at a temperature of 110 ^{o} C ± 2 ^{o} C by holding the maximum temperature for 10 hours.

Bulk densities (freely settled and tapped) and granulometry were determined after samples drying. The bulk density was determined following gravimetric method of STAS 1913/3-76 by weighting a known volume of bottom ash sample and using a gradated cylinder and an analytical balance.

Based on the results pointed out, a mechanical sieving trial was conducted using a set of sieves with the following nominal sieve opening in mm: 4; 3,15; 2,5 and 2.

The partner research team from University Politehnica Bucharest (UPB) conducted a preliminary determination of the fix carbon content and it resulted that FCC is much higher than in the lower size fractions and also in relation to the average value –table 1.

Table 1. Characteristics of initial ash and slag samples

Sample	**Size** [mm]	**Share** [%]	**FCC** [%]
PG 1	> 4	2,60	0,52
PG 2	3.15 – 4	3,14	38,82
PG 3	2.5 - 3.15	4,06	36,85
PG 4	2 - 2.5	5,82	31,27
PG 5	< 2	84,40	14,42

We collected another 150 kg of samples from the coal ash and slag stockpile belonging to Govora CHPP and, taking into account the preliminary results, we separated three fractions – see table 2.

Table 2. Characteristics of ash and slag samples

Proba	**Size [mm]**	**Share [%]**	**FCC [%]**
PG 6 	> 4	7,02	0,47
PG 7 	**2 – 4**	**16,65**	**28,44**
PG 8 	< 2	76,33	11,95

The fraction with highest FCC, respectively sample with the size between 2 and 4 mm, was named sample GI this point forward – see fig. 1.

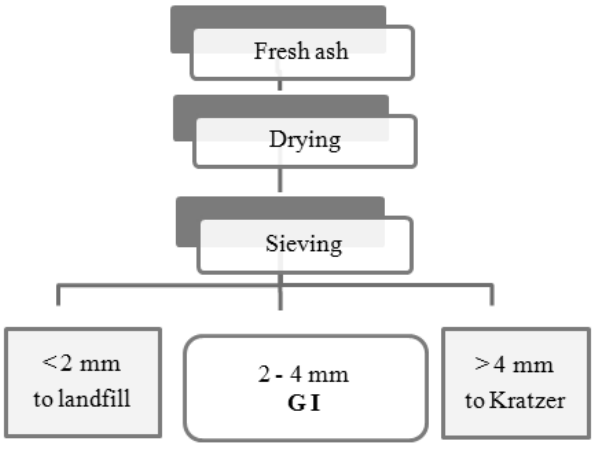

Fig.1. Procedure for initial separation

Starting from the sample GI, we developed 12 char enrichment procedures by repeatedly removing the inorganic matter through various processes such as sieving, flotation and magnetic separation.

The procedures have at least two stages of char enrichment, the first stage being sieving and dimensional separation in all cases.

The procedures were divided into three categories:

- procedures derived from dimensional separation – see fig. 2
- procedures derived from gravimetric separation – see fig. 3
- procedures derived from magnetic separation – see fig. 4

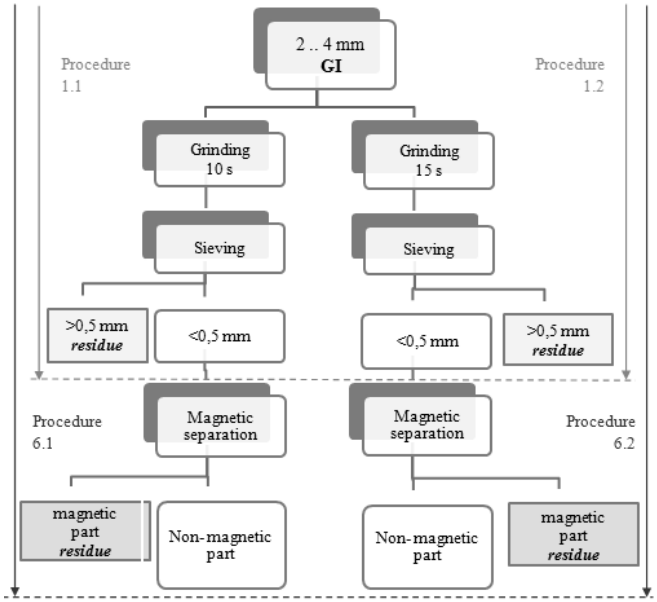

Fig. 2. Procedures derived from dimensional separation

The twelve pre-concentration procedures have been tested and, to compare the results, four parameters have been defined:

- the mass separation rate (MSR) as the ratio between the separated mass (M_{sep}) and the initial mass of the sample that is subject to the separation of a single separation procedure (M_i):

$$MSR = \frac{M_{sep}}{M_i} \quad (1)$$

- the overall separation efficiency (OSE) as the product of the mass separation yields where the initial sample is successively subjected to several separation procedures. RGS is actually the ratio of the final mass ($M_{sep,\ n}$) and the initial mass of the sample that is successively subjected to several separation procedures (M_i).

$$OSE = MSR_1 \cdot MSR_2 \cdot \ldots \cdot MSR_n = \frac{M_{sep,1}}{M_{ini}} \cdot \frac{M_{sep,2}}{M_{sep,1}} \cdot \ldots \cdot \frac{M_{sep,n}}{M_{sep,n-1}} = \frac{M_{sep,n}}{M_{ini}} \quad (2)$$

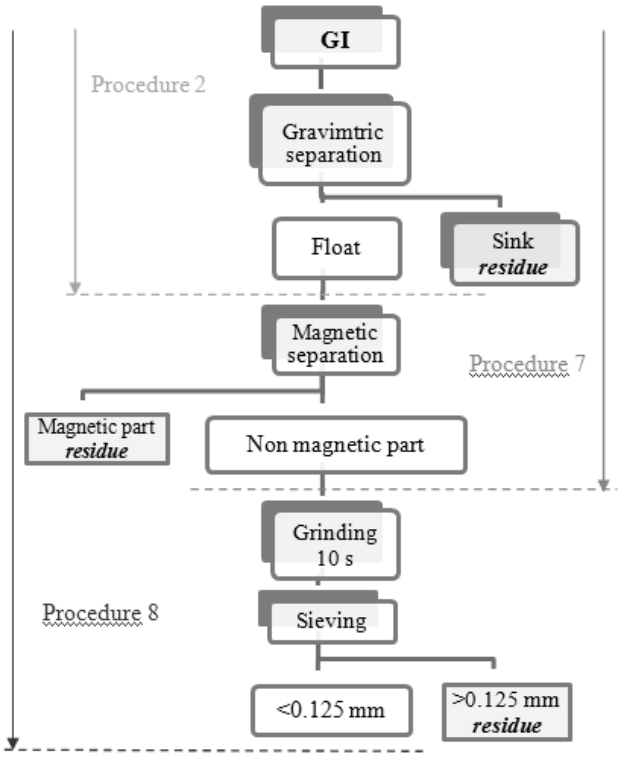

Fig. 3. Procedures derived from gravimetric separation

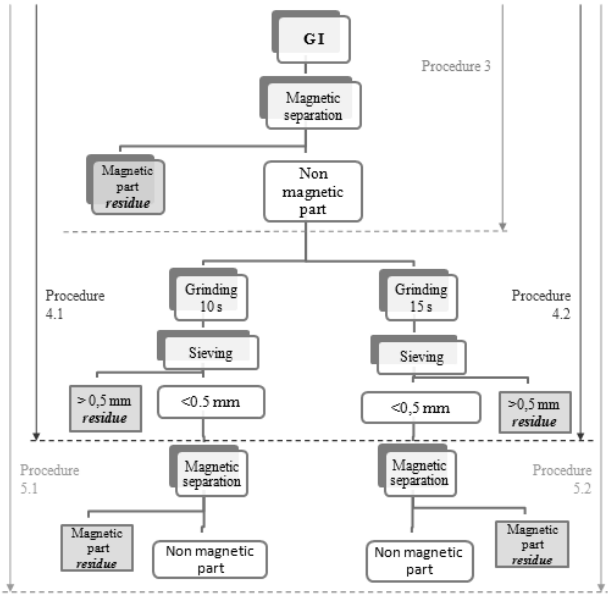

Fig.4. Procedures derived from magnetic separation

- rate of char concentration (RCC) as the ratio between the fixed carbon content in the final sample and the initial sample:

$$RCC = \frac{FCC_{fin}}{FCC_{ini}} \quad (3)$$

- rate of char recovery (RCR) as the ratio of the char mass in the final sample to the char mass in the initial sample. RCR is, in fact, the product of the mass yield of separation and the degree of concentration of char.

$$RCR = \frac{M_{char,fin}}{M_{char,ini}} = \frac{M_{sep,n} \cdot FCC_{fin}}{M_{ini} \cdot FCC_{ini}} = OSE \cdot RCC \quad (4)$$

IV. Results and Discussion

The 12 pre-concentration procedures were studied and were compared using the four parameters we previously defined.

The values of the parameters are presented in graphical form as follows:

- values of the fixed carbon content - fig. 5
- values of the overall separation efficiency - fig.6
- values of the rate of char concentration - fig. 7
- values of the rate of char recovery - fig. 8

By combining several pre-concentration procedures, the highest residual carbon concentrations exceeded 60% in the anhydrous state in two cases - procedures 7 and 8. These two procedures are the only ones that meet the requirements of the project coordinator, respectively the fixed carbon content of the final sample over 50%. Other 5 procedures lead to fixed carbon contents between 40 and 47% while the other 5 procedures result in fixed carbon contents below 33%, values close to the fixed carbon content of the GI sample (28,44%.) obtained by the initial pre-concentration.

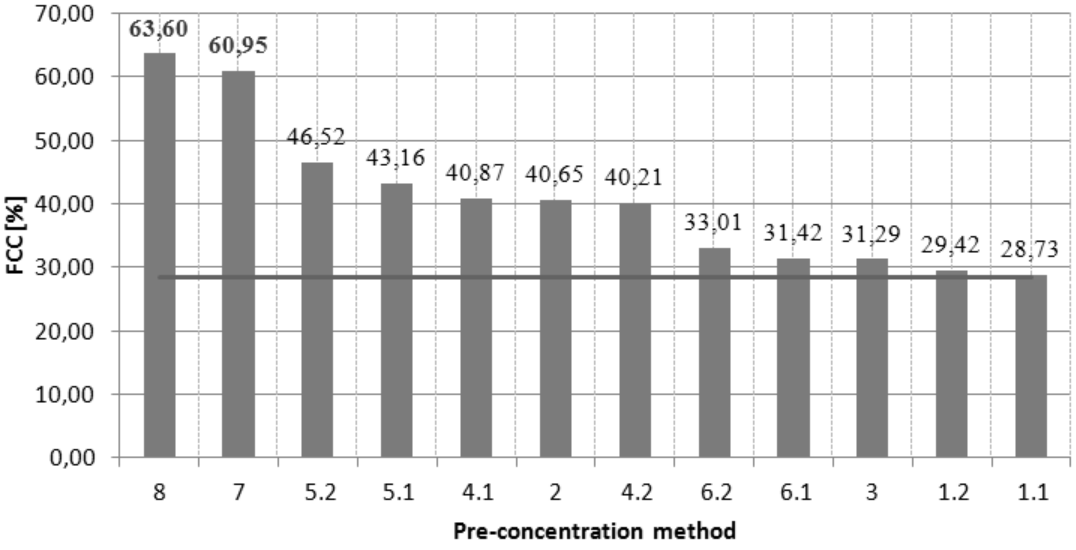

Fig.5. Values of fix carbon content (FCC) for each pre-concentration method

Analyzing the overall separation efficiency for each procedure, three procedures result in a value of over 12% and the other 9 procedures at values lower than 9.70% of the overall separation efficiency. Procedures 7 and 8, whereby large concentrations of fixed carbon are obtained, have the smallest overall separation efficiency: 4.04% - procedure 7 and 0.97% - procedure 8, respectively. The value of the overall separation efficiency is limited above the value of the initial pre-concentration procedure, of 16,65%.

By using procedures 7 and 8, the highest pre-concentration values of the char, namely 4.58% and 4.39%, are achieved. Five procedures result in pre-concentration values between 2.89 and 3.35 and the other five at values between 2.07 and 2.38, values close to the concentration obtained by the initial pre-concentration of the GI sample of 2.04.

From the point of view of the recovery of char, 8 procedures lead to values ranging from 23.8 to 28.49%. Procedure 7 leads to a value of 17.97% and Procedure 8 to a value of 4.49% for the char recovery rate.

Residual carbon (char) samples were subjected to laboratory tests to perform the technical analysis and bulk density determination, dimensional distribution of granules (granulometric composition) and calorific value.

45 technical analyzes of different samples of residual coal, 16 determinations for bulk density and for granulometric composition and 13 determinations of calorific power were performed.

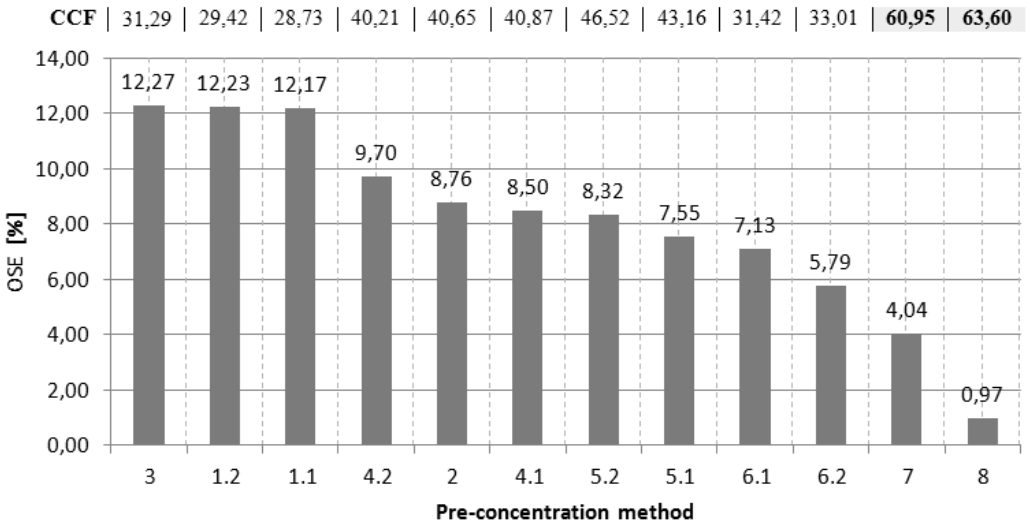

Fig. 6. Values of overall separation efficiency (OSE) for each pre-concentration method

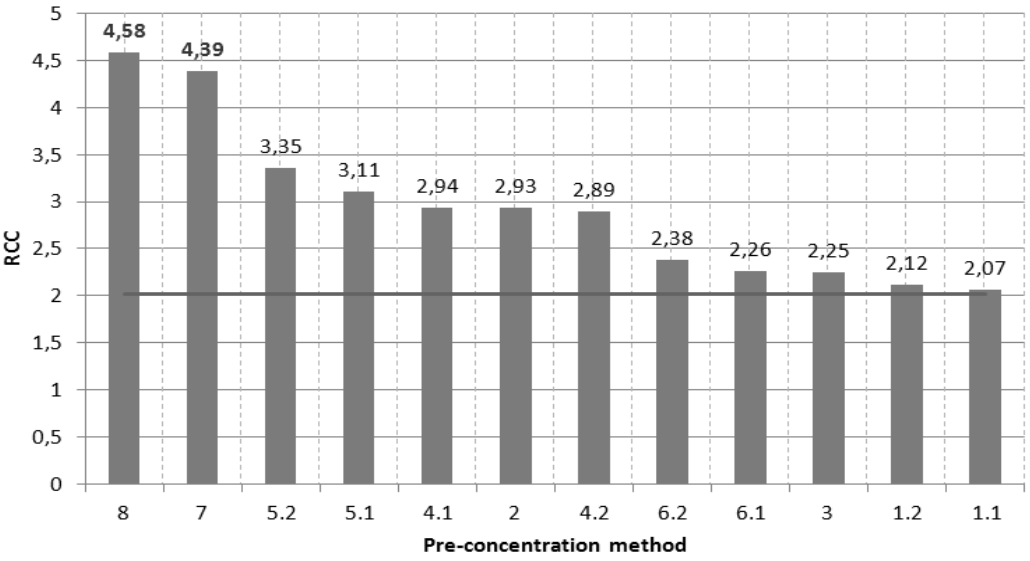

Fig. 7 Values of the rate of char concentration (RCC) for each pre-concentration method

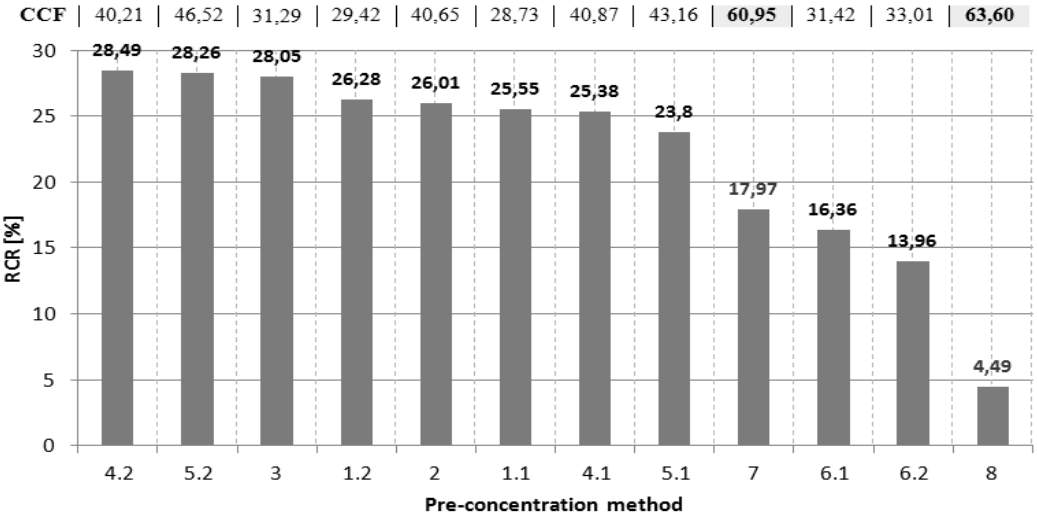

Fig. 8. Values of the rate of char recovery (RCR) for each pre-concentration method

The final pre-concentration product, obtained by applying procedure 8 and sent to the partners for graphitization, is characterized by the following average values:

- moisture 2.51%
- fixed carbon content 63.60%
- volatile content 18.33%
- ash content 17.39%
- bulk density
  - freely settled 0,407 kg / m3
  - tapped 0.586 kg / m3
- granulometric composition
  - > 0.09 mm 16.14%
  - 0,063 - 0,09 mm 17,12%
  - <0.063 mm 66.74%
- calorific power 23,14 MJ / kg

V. Conclusions

The objective was to pre-concentrate the char from slag and ash to over 50% without using substances to contaminate samples.

In order to achieve a high degree of char concentration, in the second stage, the slag and ash collected at Cogeneration Power Plant Govora, in the NOVENER project, was used. It had an average carbon content of 13.89% [9] .

Taking into account the results of the first and second step tests, the slag and ash were dimensionally sorted and the fraction 2-4 mm was used to continue the pre-concentration tests. It has the highest fixed carbon content - 28.44% at an overall separation efficiency of 16.65%. This fraction was called Govora Initial (GI).

Some pre-concentration procedures as dimensional sorting, magnetic separation and gravimetric separation, were used. Laboratory experiments showed that each pre-concentration procedure had its own efficiency limits and to obtain a higher purity of residual carbon, nine combinations of the above pre-concentration procedures were used.

Even if the individual ash particles would be completely removed from the samples, the existence of mineral matter encapsulated in the residual carbon particles prevents the further increase of the purity of the residual carbon by the procedures used. The "release" of residual carbon to such particles by grinding followed by dimensional sorting or magnetic separation has led to an increase in the fixed carbon content but also to a significant decrease in the overall separation efficiency.

Using these procedures, the UCB team obtained the increase of fixed carbon content from 13.89% to 63.60 and the decrease of the mineral mass in the tested samples from 57.57% to 17.39%, sufficient for the pre-concentration stage.

References

[1] IEA, Coal Information online data service 2017 edition
[2] Bartoňová l., Unburned carbon from coal combustion ash: An overview, Fuel Processing Technology 134 (2015) 136–158
[3] Cruceru Mihai et al, Concentration of the residual carbon from bottom ash by particle size separation, International Journal of Geology, vol. 11, pp. 1-7, 2017
[4] Predeanu, Georgeta, et al. Coal ash characteristics to assess its possible uses. *International Multidisciplinary Scientific GeoConference: SGEM: Surveying Geology & mining Ecology Management*, 2013, 309.
[5] Universitatea ”Constantin Brâncuşi” din Târgu Jiu - Proiect LIFE 10/ENV 729 RO, Anexa tehnică 4.5, 2013
[6] CHARPHITE Project Poposal“Coal char as a substituting material of natural graphite in green energy technologies” “Third ERA-MIN Joint Call (2015) on Sustainable Supply of Raw Materials in Europe”.
[7] Cruceru, Bruno Valentim, Georgeta Predeanu, Traian Abagiu, Valerica Slăvescu, Lucica Anghelescu Increasing the residual carbon content from bottom ash by particle size separation, WSEAS TRANSACTIONS on ENVIRONMENT and DEVELOPMENT, Volume 12, pp. 337-342, 2016
[8] Popescu, Luminita Georgeta, et al. Analysis of heavy metal content to evaluate leaching characteristics of coal ash wastes. *International Multidisciplinary Scientific GeoConference: SGEM: Surveying Geology & mining Ecology Management*, 2013, 1: 33.
[9] "Energy conservation innovative coal-fired technology-NOVENER project" Innovation project coal burning technology SCIENTIFIC AND TECHNICAL REPORT