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Flue Gas Cleanup at Temperatures about 1400 °C for a Coal Fired Combined Cycle Power Plant: State and Perspectives in the Pressurized Pulverized Coal Combustion (PPCC) Project

Keywords: Combined Cycle, Coal, Flue Gas Cleaning, PPCC (DKSF)

1 Introduction

The PPCC technology, a combined cycle, requires comprehensive cleaning of the flue gases because coal contains a large variety of minerals and other substances. This would lead to fast destruction of the gas turbine blades due to erosion and corrosion. The present specifications of the turbine manufacturers for the required flue gas quality are at a maximum particulate content of 5 mg/m^3 s.t.p., diameter of < 5 μm, and a maximum alkali content < 0.01 mg/m^3 s.t.p. The PPCC project is aimed at cleaning the flue gases of pressurized coal combustion. This method will be applied at temperature ranges where the ash is in a liquid state and which will be thus cleaned from coarse particulate material by agglomeration and inertial force separators. Appropriate separating methods are also being investigated and developed for the hazardous gaseous contents, e.g. alkali compounds, which are released during the coal combustion process. The following companies are working on the development within the scope of a collaborative project to find a feasible technical solution: Babcock-Borsig-Power Env. GmbH (BBP Env.), E.ON Kraftwerke GmbH, SaarEnergie GmbH, Siemens AG, and Steag AG.

The questions pursued within the scope of this research projects pertain to the following issues:

- combustion of pulverized coal under pressure,
- extensive removal of liquid ash from the flue gases,
- removal of gas contents such as alkalis, which are hazardous to the turbine,
- selection of suitable materials for the high temperature range,
- clarification of turbine - relevant questions,
- testing and development of adequate measuring methods.

With a focus on process development a testing plant with a thermal output of 1 MW has been erected in Dorsten in 1991. It can be operated using pressures up to 18 bar and a furnace temperature up to 1750 °C.

2 Objective

Power and heat generation from coal will remain indispensable, even in the future, considering the resources on our planet. A high efficiency is a determining factor when producing electrical energy due to the limited availability of fossil fuels and considering the necessity to reduce the pollution of our environment.

In conventional coal-fired power plants with a pure steam turbine process, projects are presently being implemented allowing net efficiency rates around 45 %. Efficiency rates up to 60 % can be achieved with the combined cycle power plant technology. In these combined gas-steam cycle processes only ash-free fuels like natural gas and light fuel oil can be presently employed. Past investigations aimed at implementing a gas-steam cycle process by gasifying coal have shown that, due to the costly and complex coal conversion method, the efficiency drops to such a low rate that this method offers no substantial advantages compared to a modern steam turbine process. Therefore, from today's point of view, the pressurized combustion of coal with an efficient ash and alkali separating rate can be considered the most suitable method to employ hard coal in the combined gas-steam turbine process with an efficiency of approximately 55 % (figure 1) (BMWi 1999; Heyn 1999).

3 Project Pressurized Pulverized Coal Combustion (PPCC)

The PPCC -technology requires a comprehensive cleaning of the flue gases because coal contains a large variety of minerals and other substances. These lead to the fast destruction of the gas turbine blades due to erosion and corrosion. The present specifications of the turbine manufacturers for the required flue gas quality are at a maximum particulate content of ≤ 5 mg/m^3 s.t.p., a diameter of < 5 μm, and a maximum alkali content < 0.01 mg/m^3 s.t.p.

The investigated method is aimed at cleaning the flue gases of the coal, which is burned under pressure. This method will be applied at temperature ranges, where the ash is in a liquid state, and which thus will be cleaned from coarse particulate material by agglomeration and inertial force separators. Appropriate separating methods are also being investigated and developed for the hazardous gaseous contents, as e.g. alkali compounds, which are released during the coal-combustion process. The basic design of a plant with Pressurized Pulverized Coal Combustion is illustrated in figure 2.

To find a suitable technical solution, the following companies work on the development of the Pressurized Pulverized Coal Combustion Process within the scope of a collaborative project: Babcock-Borsig-Power Env. GmbH (BBP Env.), Eon Kraftwerke GmbH, Lurgi Envirotherm GmbH (until June 1999), SaarEnergie GmbH, Siemens AG, and Steag AG.

3.1. Priorities

The questions pursued within the scope of this research project pertain to the following issues:

- Combustion of pulverized coal under pressure,
- Extensive removal of liquid ash from the flue gases,
- Removal of gas contents such as alkalis, which are hazardous to the turbine,
- Selection of suitable materials for the high-temperature range,
- Clarification of turbine-relevant questions,
- Testing and development of suitable measuring methods.

Tasks and subject matters were distributed among the different partners depending on their field of expertise (figure 3).

Furthermore, as participants in the "Druckflamm" program, a large number of universities and research institutes are involved in basic research in connection with the application of future technologies, as e.g. combustion under pressure, behavior of compound substances under the influence of high temperatures and pressures, behavior of ceramics, electrical effects, and measuring methods. The German Federal Department of Trade, Industry, and Technology (BMWi) subsidizes the project with a share of 50 % (FKZ 0326840).

4 Status

After initially experiencing material problems with ceramics exposed to the liquid slag during the testing operations, the subsequent operations were mainly targeted at running tests in connection with particulate separation. The aim was to determine separating parameters, which are essential for the development of a commercially operated plant.

4.1. Test plant

After considerably optimizing the availability and performance of all components, the test runs of the completed phases show that all components are basically suitable for the process. In numerous steps, the operating safety and the stability were substantially improved. Up to now, deficiencies and limitations have mainly been caused by conventional components. Approaches towards further optimizations (in detail and generally) could be revealed and are now being implemented. New "tools", like the possibility for numeric simulation, but also the worldwide access to information promise a progressive development of components and materials in the future. In detail, the measures apply to pressure burners, furnace, apportioning of coal, and instrumentation technology.

4.2. Separation of liquid ash by means of packed-beds

In November 1998, a first outline was presented on the process, the PPCC-testing plant in Dorsten, its operation and on the state of flue gas cleaning (Druckflamm 1998). At that time, the fact that packed-beds allowed a safe particulate separation with a diameter of > 5 μm was already established, as SEM pictures of fly ash samples and LDA-measurements verified. The remaining particulate concentration is, however, still too high for the operation of a gas-turbine.

The investigation of different bed arrangements showed that a sloping separator, in which the collected slag moves downwards within the bed, has the positive consequence of thus removing the liquid from the passing gas stream. However, the flue gas cleaning process is negatively affected by the resulting wear on the material and a subsiding of the balls in the upper section, which results in the formation of flue gas leaks. A vertical, U-shaped separator proved to be a practical solution for the separation of slag. No further problems occurred during the test runs. The bed-material utilized in the separator consisted of balls, because this design is accessible for use in computations and modelling. The advantage for the plant operation is the fact that balls can be produced from different materials, and material wear will result in sagging and a re-filling of the balls could follow. It was furthermore determined that separating systems, which are based on inertial force, will allow a further particulate reduction only in connection with considerable expenses (high velocities and therefore an increasing material wear, a considerable pressure loss and thus a reduction of the efficiency). Tests of this nature did not result in any significant particulate decrease. Therefore a second separating stage was installed aimed at the reduction of the remaining superfine particulate (d < 5 μm) content.

4.3. Separation of superfine particles

The following methods are available for the separation of superfine particles:

Separation by means of demister units: To allow separation of droplets in the aerosol section, multi-layer mesh or network made of wires or fibers with a distance smaller than 1 mm are used (e.g. behind scrubbers). If such a mesh is made of an inert, temperature-resistant material, it can also be employed under PPCC-process conditions. A test run employing a mesh of platinum wire (Fp 1720 °C), however, showed, that this type of separator exhibits a spectrum of new difficulties. The slag, e.g., has a high affinity to the platinum wire thus adhering there and filling the spacing between the wires instead of flowing off from the mesh. As a consequence the adhering slag exhibits a loss of alkali compounds with the result of an increasing melting point. Regarding costs of the material and recovering it after a test run, no further experiments with a device like that are planned.

Separation in accordance with the ESP principle: A separation based on the classical ESP principle has also been considered. It will, however, be pursued no longer, because a corona cannot form on the spray electrodes due to the proven electrical conductivity of the flue gas at process temperatures. Charges that form on the particles are directly emitted via the flue gas. The possibility of using the effect of an ”electric wind”, an interpretation referring to the mechanism of the ESP-separation, will be further investigated, because a flow of charged particles (electrons and ions) between electrodes in the hot flue gas may result in a transportation – and therefore also in a separation activity.

Separation by utilizing plasma effects occurring due to the existing process temperatures: A completely new aspect, which might elucidate the reasons for the remaining size of the particles, as well as for the problems relating to their separation, is revealed by considering the electrical properties of the flue gas. The flue gas, including its contents, can be considered a cold-plasma at the existing process temperatures. This means that the flue gas behaves macroscopically neutral. But within the flue gas, a separation of charges occurs. This fact has been verified by the conductivity of the flue gas, as well as by the charge of the particles (Kleinwechter et. al. 1999).

The charges on the particles cause a repulsion, an effect which will become especially noticeable on smaller particles. The result is that these particles cannot agglomerate any longer and can therefore not be separated. A local interference of the electrical properties could be one way of causing a separation. This can, e.g., occur via surfaces which get into contact with the flue gas. Therefore it appears to be a promising concept to employ conductive materials (platinum wire, materials containing ZrO_2) which can carry off the charges, thus causing a local change of the charge. Designing a local electrical gradient by combining suitable ceramic materials is the subject of intensive research.

It is furthermore considered to utilize the observed effect that increased slagging reoccurs in cooler sections of the flue gas path (resulting from the changes in the properties of the cold-plasma) in a targeted way.

4.4. Particulate content determination

Before naming and discussing particulate contents, it should be illustrated how these contents are determined and what a given value reflects. The measurements are carried out by means of a retractable measuring probe, which is inserted via a lock system into the plant behind the flue gas cleaning stage. The stub consists of an outer, water-cooled steel tube, into which a heatable quartz measuring head has been installed. The flue gas path is sampled isokinetically and after passing the quartz tube, it flows through a fly ash collecting filter (figure 5). A reference measurement at room temperature with a measuring device according VDI-directive 2066 resulted in a satisfactory compliance of the determined values (deviation approx. 5 %). The measuring system operates reliably and supplies repeatable values.

This type of sampling requires a flue gas withdrawal at approx. 1200 °C, a cooling to approx. 700 °C during the passage through the quartz stub and a particulate collection on a filter at approx. 650 °C. At this temperature range, the conversion of some oxides into sulfates ($CaO \longrightarrow CaSO_4$, e.g.) occurs. Out of this gaseous phase, a number of sulfates condense (composite sulfates, sodium and potassium sulfates). Aside from the primary particles, these composites are also recorded during the weighing process of the fly ash sample and appear as a part of the fly ash content. A rough estimate of the condensate content can be determined by means of a thermogravimetrical analysis. Such an analysis, performed on a fly ash collecting filter from a sampling, in the course of which a content of 350 mg/m^3 s.t.p. was determined, revealed that about 150 mg have to be substracted due to evaporation during the heating-up phase starting at a temperature of 600 °C (figure 6) (Enders and Putnis 1998). This condensate content in the sample should be taken into consideration during the analysis of the particulate content, prior to a discussion of the state of the effectiveness of particulate separation.

4.5. Alkali reduction

Up to the present, no test runs have been carried out during which alkali reduction equipment was employed. Previous investigations were directed at the behavior of slag to alkalis, as well as the behavior of materials designed to bind alkali compounds (Witthohn et. al. 1998). A test series in the course of which alkali compounds were added to fuel showed that slag has a distinct buffer capacity for alkalis, which can be utilized further. By introducing additional steps, the remaining content can be reduced to achieve turbine compatibility:

- Lab tests with radioactive-labeled alkali salts showed an adequate rate of retention by getter-materials (Hennig et al. 2000);

- Retention by getter-materials (and by slag) in an electrical field also appears promising.

4.6. Materials: in particular Ceramics

During the previous test runs in Dorsten it became obvious that

- font-molded and isostatically compressed ceramics with a high Cr_2O_3-content and a dense surface proved a success and
- that the available materials allow the development of the process.

However, ceramics containing chrome oxide tend towards a vaporization of the chrome in the water-steam containing flue gas (Karwath et. al. 1999), which can consequently be retraced in the fly ash samples. This becomes obvious due to the intensive coloring of the collected material.

Apart from the condensates, the chrome oxide content, which does not originate from the fuel, should also be considered when discussing the particulate content, especially when it reaches a range of 10 – 20 %.

In spite of a rather satisfactory behavior of ceramic materials in the test plant, it is still necessary to give special attention to this field. On one hand, the release of chrome must be reduced, and on the other hand a service life must be obtained, which allows the operation of large-scale plants with their usual inspection intervals. First samples from materials indicate the approaches are promising.

Special attention was given to the graining, the homogeneity, and the purity of the raw material during production and to the designing process in view of an optimum application. Materials, which have been furnished with a new surface coat thanks to new surface coating methods, give reason to expect if not a complete, at least a substantial reduction of the chrome content in materials.

5 Current tests

Tests on superfine particulate separation by utilizing low-temperature plasma effects have been carried out with material composites. For this purpose, a packed bed was composed, in which materials from ceramics containing zirconia and chrome oxide were combined and arranged in rows. The flue gas passed through this bed at a velocity of about 0.5 cm/s. Compared to a bed consisting of ceramics, which contain chrome oxide or aluminum oxide only, and compared to a separator made of foam-ceramics – which can be employed in areas without any problems, where the flue gas contains only superfine particles – this arrangement displays deposits on the ceramic material, which consist of separated slag droplets (figure 7).

Due to this arrangement, the particulate content could be reduced from approx. 350 mg/m^3 s.t.p. to a range lower than 200 mg/m^3 s.t.p. without any attempts to optimize this arrangement. This is visibly indicated by the deposits in the probes of the fly ash collecting device, which consist of superfine ash only (figure 8).

Having in mind the above comments on condensate contents, which can be regarded as constant under the same conditions, this is a clear indication for a considerable progress in the field of particulate cleaning (figure 9).

Apart from this passive method of particulate separation, laboratory tests have shown an additional possibility, where the electrical properties of the flue gas are also utilized. In this case, a gas flow with particles in the submicrometer range was conducted at a temperature complying with the temperature of the process gas in the test-plant through an electrical field which was applied on the outside. 98 % of the particles were separated (figure 10) (Weber et al. 2000). A modified setup is under test in the plant in Dorsten. The first results show a significant effect on the particle content as well as on alkali extraction into the liquid slag. A deeper basic knowledge, however, of the effects implied has still to be reached to construct an optimized aggregate.

6 Future Activities

The present findings result in the necessity to deal with the following issues for the development of the process:

- Formation of particles
  - Influence of the coal mineralogy on the formation of particles.
  - Impact of the flame temperature on the formation of particles, influences of operational parameters on the formation of particles (pressure, temperature level, furnace, air heating).
- Particulate separation
  - Possibilities and limitations for the separation of superfine particles by means of mechanical separators.
  - Possibilities for the utilization of plasma effects: Local discharging of particles by means of suitable materials (in view of the desired temperature range) for a deposition or aggregation.
- Reduction of the flue gas contents with corrosive effects.
- Ceramics with a creep resistance limit in the field of particulate separation.
- On-line measuring methods suitable for process conditions for the required maximum concentrations.

7 Summary

At the present time, we can summarize as follows:

- A further reduction of the clean gas particulate content, especially in the problematic range of d < 5 μm, is feasible and promising considering different approaches with different possibilities.
- A reduction of the alkali-compounds to the required values is feasible according to present model calculations. There are different options relating to the implementation.

For this purpose, it will be necessary to obtain relevant fundamental research data, which will allow an optimization of the individual cleaning steps. With the existing plant, no optimizing tests are supposed to be run at the time, but, in principle, the intention is to show that certain measures have a significant influence on the gas-cleaning process (particulate separation, alkali reduction). It has been shown that certain problems can only be revealed through the operation of the PPCC-plant, which cannot be identified or handled with any other plant. Therefore it will be necessary to operate this plant further with the following priorities:

- Reduction of particles < 5 μm,
- Alkali reduction,
- Material creep resistance limit,
- Development of components.

Outlook

It can be assumed that, in the course of the current project phase (until the end of 2002)

- the dust values can be reduced to about 20 mg/m^3 s.t.p. According to the turbine manufacturers, 5 mg/m^3 s.t.p. are permissible.
- The alkali concentration will be reduced to about 0.05 mg/m^3 s.t.p. In this case, the limit value ranges around 0.01 mg/m^3 s.t.p.

By the end of the testing phase it can be assumed that the above-mentioned values can be maintained over a period of several days. The past results and successes in superfine dust separation render us confident that the flue gas from coal combustion can be cleaned to an extent where it meets the requirements of a gas-turbine. Therefore we believe that the PPCC-process is feasible.

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9 Figures

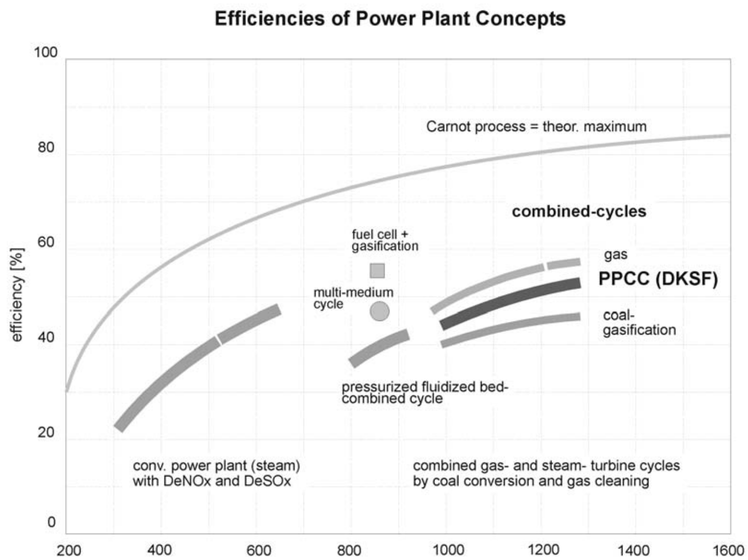

Figure 1: Upper process temperature / gas turbine inlet temperature, respectively (ISO, °C)

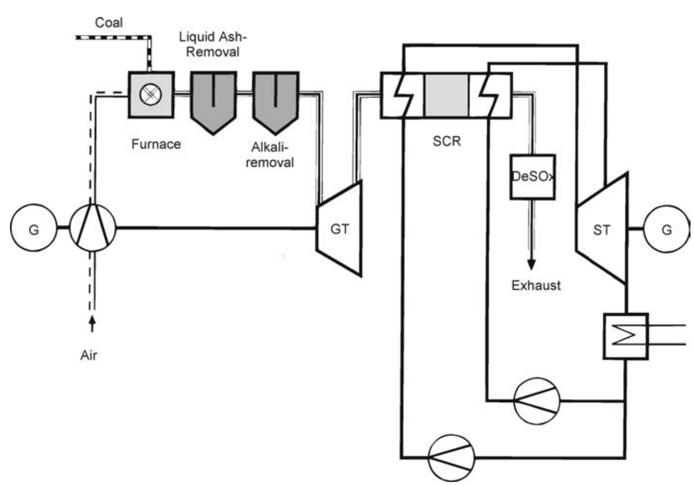

Figure 2: Basic layout sketch of the combined gas-steam turbine cycle with Pressurized Pulverized Coal Combustion (PPCC)

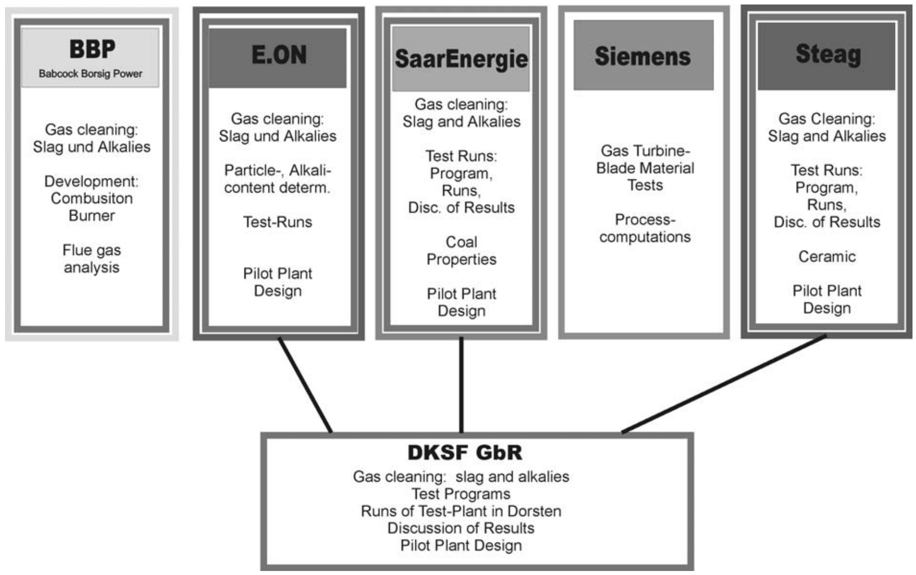

Figure 3: Partners and Tasks

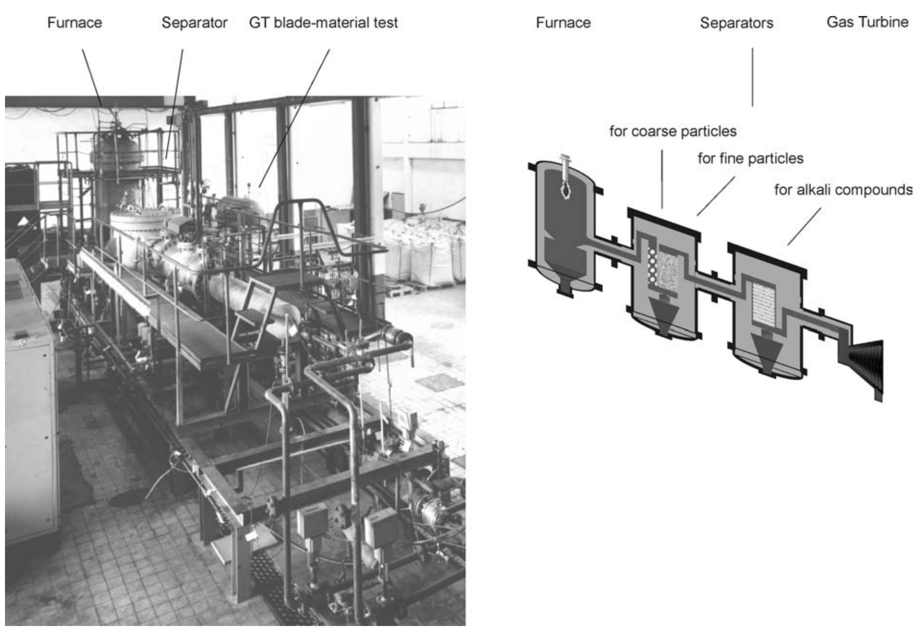

Figure 4: 1 MWth PPCC-testing plant in Dorsten including the components to be developed: furnace, liquid ash separator, alkali separator, gas-turbine material testing facilities

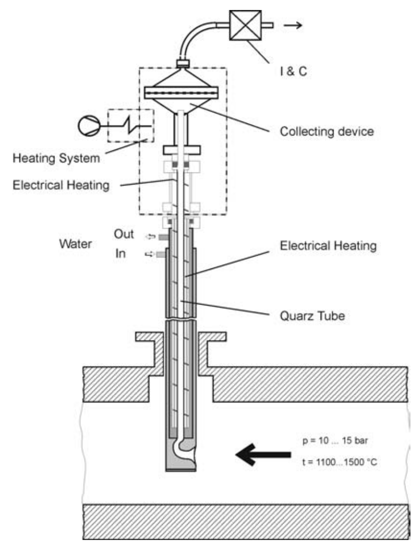

Figure 5: Basic layout sketch of the fly ash collecting device

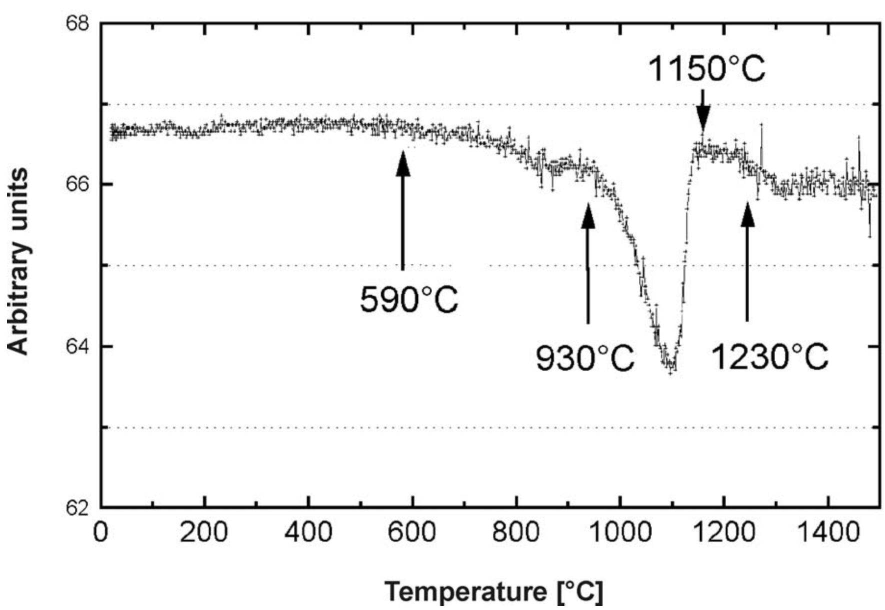

Figure 6: Measuring report of a DTG-analysis performed on a fly ash sample

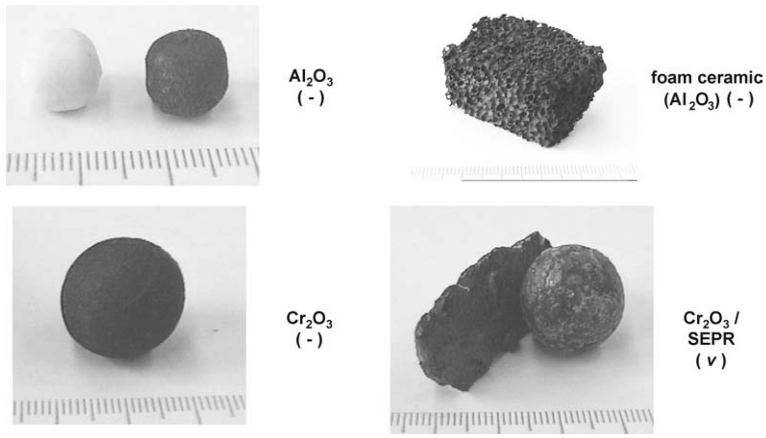

Figure 7: Different behavior of ceramics in contact with superfine ash in the process temperature range: Only the combination of special ceramics leads to a separation of superfine dust (lr):
alumina oxide (ul), chrome oxide (ll), foamed ceramics (Al_2O_3 (ur)), combination of ceramics containing zirconia / chrome oxide (lr)

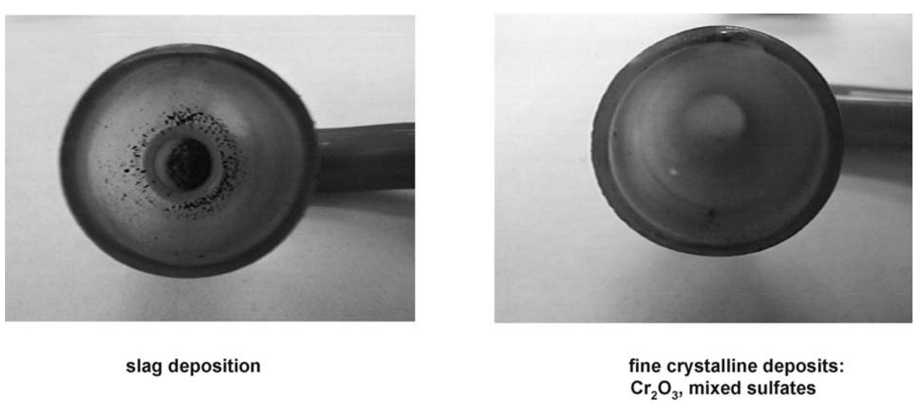

Figure 8: View of inlet probe heads: Comparison without (1) / with superfine ash separation (r)

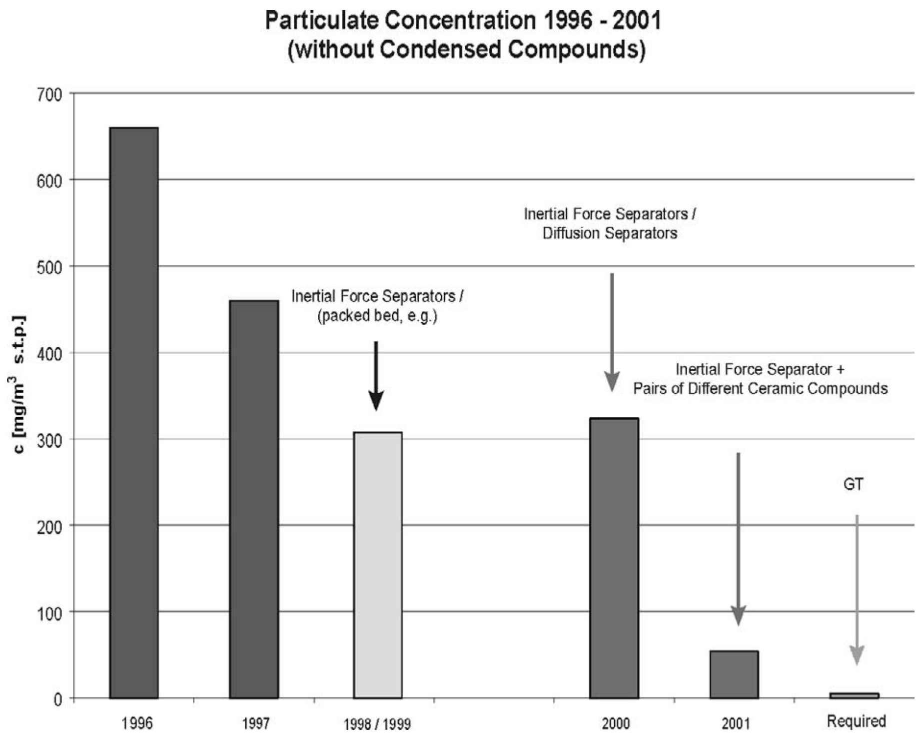

Figure 9: Particulate contents after flue gas cleaning with different separating mechanisms: The additional introduction of a separating stage for particles < 5 μm results in a considerable improvement of the flue gas quality

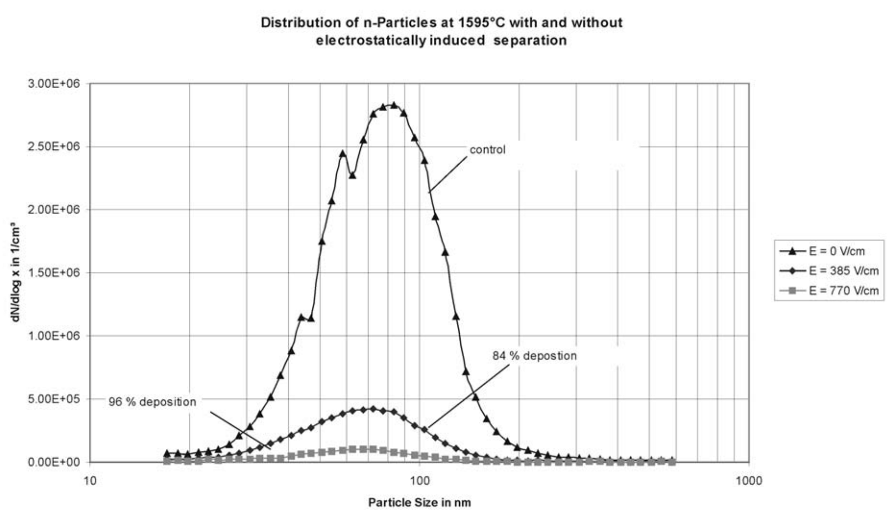

Figure 10: Separation of submicro-particles in an electrical field at high temperatures