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Use of Calcareous Fly Ash in Germany

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Abstract

In German lignite fired power stations about 10 million tonnes of ashes and 5 million tonnes of flue gas desulphurisation gypsum are produced every year. Most of the ashes are used for the refilling and reclamation of depleted opencast lignite mines. Furthermore, they are used in underground mining, for surface recultivation, soil beneficiation, cement production and as addition to concrete. FGD gypsum is used in the gypsum and cement industry and increasingly as fertilizer and soil conditioner.

The utilisation of the calcareous fly ash from lignite fired power plants is depending on their chemical, mineralogical and physical properties. These properties are influenced by the power plant technology, the source of coal as well as the type of coal feed. A constant product quality is of greatest importance for utilisation in hydraulic binders, cement and concrete.

The paper deals with the utilisation of calcareous fly ash from lignite fired power stations in Germany, existing fields of application and results of recent research work regarding their utilisation in blended cement.

Keywords: lignite, calcareous fly ash, properties, reactivity, utilisation, reclamation, mine fill, soil beneficiation, blended cement, concrete

1. Introduction

In Germany, the share of energy and steam production by coal in 2010 amounted to 42.4 %. In 2010, about 44 million tonnes of coal and about 150 million tonnes of lignite were burned in coal and lignite fired power plants. By this, about 22 million tonnes of coal combustion products (CCPs) were produced including about 9 million tonnes of fly ash and about 5.5 million tonnes of FGD gypsum from lignite. The utilisation of fly ash is influenced considerably by their quality, i.e. primarily by the fluctuations of the chemical and physical properties. Regarding this, coal fly ash has more favourable prerequisites than most lignite coal ashes whose composition is subject to comparatively larger fluctuations. FGD gypsum is nearly completely used as a replacement for natural gypsum in the gypsum and cement industry as well as for agriculture.

Lignite fly ashes are predominantly used for refilling opencast mines, partly after treatment with water or water from flue gas desulphurisation (FGD water) in order to form stable hydration products. Furthermore, the ashes are used in underground mining, for surface restoration, soil beneficiation, immobilisation purposes as well as for cement and concrete production. For the use as concrete addition a technical approval was issued, which shows that the properties of the fly ash complies with the requirements of DIN EN 450 "Fly ash for concrete". The recent research work focussed the use in blended cement production. Fly ash was also used in pilot projects for hydraulic road binders and for blended cement CEM II-B (W-LL).

2. Lignite mines in Germany

In Germany, mining of lignite is concentrated in four regions where lignite is exclusively extracted in opencast mines (see figure 1). In 2010 in total 169.4 million tonnes of lignite were mined in the Rhenish area in the western part of Germany near the town-triangle Köln-Aachen-Mönchengladbach (54 % by mass), the Central German area near Leipzig (12 % by mass), the Lausitz area in southeast of Brandenburg and northeast of Saxony (33 % by mass) and the Hirschberg mine near Helmstedt (1 % by mass) [1].

Most of this lignite is burned in big power stations nearby the mines, lignite dust and briquettes are transported by train or trucks to customers.

3. Production of CCPs in lignite fired power stations

CCPs in lignite fired power stations cover mostly fly ash, bottom ash and FGD gypsum from flue gas desulphurisation of dry bottom boilers (see figure 2). In most of the lignite power plants pulverized fuel is used. Lignite is ground to fine dust in coal mills and is pneumatically fed to dust burners. In the furnace of the power plant the pulverised lignite is combusted. The heat produced heats the water in the water-steam-circuit, the developing steam powers the turbine. A minor part of the mineral matter from the lignite falls down to the bottom of the furnace where it is removed as bottom ash in a water bath. The major part of the ash of approximately 80 % of the mineral matter is carried along with the flue gases to the electrostatic precipitator. The ratio of bottom ash to fly ash volumes produced in lignite power plants is roughly 1 to 4. After dust separation in the electrostatic precipitator (ESP) the sulphur is removed from the flue gas in the flue gas desulphurisation (FGD) unit, where FGD gypsum is produced. The clean gas, free from dust and sulphur is carried off via the chimney or the cooling tower.

In table 1 the statistical figures regarding origin and utilisation of CCPs in Germany in the year 2010 for lignite fired power plants are listed [2]. In total, approximately 15 million tonnes of CCPs were produced including 8.5 million tonnes of lignite fly ash, 1.6 million tonnes of bottom ash and 4.8 million tonnes of FGD-gypsum.

4. Utilisation of lignite fly ash

The utilisation of fly ash is influenced considerably by their quality, i.e. by the fluctuations of the chemical and mineralogical composition as well as the physical properties. Regarding this, coal fly ash has more favourable prerequisites than most lignite fly ashes whose composition is subject to comparatively larger fluctuations. The fluctuations of chemical parameters of lignite fly ashes from German power plants in different mining areas are given in table 2 [3]. It has to be considered that the fluctuations of fly ash from single power stations are much smaller.

The biggest fluctuations are observed with SiO_2 which is caused by sand layers in lignite seams. Fly ashes of the Rhenish and the Central German area show high amounts of lime and sulphur compared to those from the Lusatian area. The amounts of reactive silica, lime, free lime, magnesium oxide, aluminium oxide and sulphur, as well as amorphous material, are markers for reactivity and also for potential fields of utilisation.

In 2010, about 97 % by mass of the fly ash from lignite combustion is used for the refilling and reclamation of depleted opencast lignite mines [2]. The remaining 3 % were used in underground mining, for surface restoration, for soil beneficiation, for immobilization and for production of cement and concrete. The prerequisites for some of these application areas as well as existing limit values in standards or regulations will be described below. In addition, a specific project for soil beneficiation as well as the results of research and pilot projects for fly ash in hydraulic bound roads and for cement production (CEM II-W) will be presented.

4.1 Refilling of depleted opencast lignite mines

Based on the German mining regulation [4] the depleted opencast mining areas have to be refilled, or at least partly refilled, for agricultural and forestry purposes. As this law is strictly binding the largest share of the ash is used to reduce the mass deficit produced by lignite mining. For this, the ashes have to fulfil the requirements of the Landfill Directive [5].

Refilling opencast mines is practiced in different ways in the mining areas of Germany. In the Central German area water treated fly ash is refilled. Water treatment is performed to slake the free lime which could cause unsoundness of compacted layers in contact with water. The self-hardening properties of the fly ash form stable monoliths over time. In the Lusatian area moistened fly ash is used for refill, partly moistened with water from flue gas desulphurisation (FGD water). In the Rhenish area fly ash is mixed with FDG water to provide a good basis for stable and long-term resistant bodies. Depending on their origin lignite fly ashes have different self-hardening properties. During the complex process of hardening and further hydration new minerals grow in porous structures leading to a reduced pore volume, increasing density and strength of the structure and hence to reduced water permeability.

In dependence of the application, i.e. bank stabilization, safety measures or refill of mines, the mix of fly ash and FGD water has to meet special requirements regarding compressive strength, water penetration and leaching. The refill of mines is monitored by the local mining authorities. Materials used for this purpose are defined in the mining and restructuring plan of the mining company and need to be approved by the authorities [6].

4.2 Infill of underground mines

In underground mining lignite fly ash is used as a constituent of binders or directly as sealing and stowing material. Mining mortars are predominantly used for consolidating the roadway supports and backfilling of mine galleries. They also serve for building up load-bearing and solid edge dams as well for stowing and damming abandoned mines. Due to their self-hardening properties they are also used directly for underground mine fill. For this, a slurry is pumped to the underground galleries. After setting and hardening the strength formation is controlled.

The use of lignite fly ash in underground mining has to follow the mining regulation and the approval of the responsible mining authorities. The requirements to be met are different in each country.

4.3 Soil beneficiation

Lignite fly ash is an effective agent for chemical and/or mechanical stabilization of soils. This soil beneficiation is of special importance in road and railway construction. Fly ashes rich in lime are effective for stabilizing soils, due to the amount of free calcium oxide. Those with low lime content can be used in combination with cement or lime for the same purpose.

Sand and gravel with small grain size distribution are beneficiated with fly ash to increase the fine fraction smaller than 0.063 mm. Clay soils are treated to mitigate the volumetric changes of the clay soil due to fluctuating moisture content by binding the soil particles into a fixed matrix. Another application of lignite fly ash is the stabilization or strengthening of a soil, such as the treatment of soil to improve its bearing capacity, the stabilization of backfill to reduce lateral earth pressure, and the stabilization of embankments to improve slope stability.

Over the last decade lignite fly ash from Central German area was used for soil beneficiation for railway and road construction [7]. In the north of Germany, a highway is constructed on clayey and wet ground, the A72 between Rathendorf and Frohburg. For the earthworks 1.5 million m³ had to be moved and 2 million m² to be beneficiated. The ground consists of 90 % clay with up to 30 % water content. For the beneficiation a minimum binder content of 3 % was proposed. For the binder material extensive tests for alternative binders including lignite fly ash have been performed. Compared to other road binders fly ash turned out to developed a strength of more than 12.5 MPA but showed a better water fixation than HRB of comparative strength classes. After discussion with the road authority the soil was beneficiated in a two-step process with lignite fly ash. Beside the technical parameters the impact of lignite fly ash on soil and ground had to be assessed. The A72, Los 3.1, is the biggest ash construction project with more than 120.000 t of lignite fly ash [8].

4.4 Production of blended cement

Actually, no blended cement with lignite fly ash is produced in Germany. Experiences with lignite fly ash cement were gained in the past in the former GDR and recently in new research projects.

In 1968, a blended cement named PUZ 225 was produced by cement works Rüdersdorf. A fly ash rich in silicon and alumina was used, the sulphur content was 8 % by mass. The fly ash content was about 40 % by mass of the cement. The cement showed a similar behaviour regarding heat treatment, frost-resistance and corrosion as normal cement. In 1984, in the same cement works a cement with lignite fly ash and low alkali content PZ 9/35 was produced. The fly ash ratio was 20 to 25 % by mass. Cement PZ 9/40a, produced in the 1970ies as a successor of PZ 9/35 and aiming in compressive strength of more than 40 MPa, was preferably used for production of concrete products, because of his high compressive strength after heat treatment [9].

As the cement industry has to further reduce CO_2 emissions from their production process and as the technical optimisation is exhausted further CO_2-reduction can only be reached with the higher production of blended cement. By this, also lignite fly ash may be used for cement production if the requirements for siliceous and calcareous fly ash for the production of blended cement as defined in DIN EN 197-1 [10] are met.

In the early 1990ies, the properties and experience in utilisation of lignite fly ash was subject of extensive research work [11]. In close follow a project was organised by the cement industry for the use of calcareous fly ash in the cement industry [12]. They have tested calcareous fly ash from different power plants for their use in blended cement. It was concluded that the requirements of EN 197-1 [10] are normally not met. However, the ashes contributed to strength formation. To optimise the beneficial use the ashes should be milled to increase the fineness. Due to the free lime and periclas content soundness should be tested also with cold water storage test. In the following years a project for technical and ecological optimised cements was performed [13, 14]. The results showed that although calcareous ash does not meet the requirements in EN 197-1 they could be used for cement production.

Also the power industry organised research project on the characteristics and performance in blended cement of fly ashes from six power stations of different mining areas (Rhenish, Central German and

Lausitz area) and different capacity focussing the homogeneity of the ashes. Within six months 19 samples were taken from each power station representing daily, weekly or monthly taken samples. The samples were characterized by testing of physical and chemical parameters. Furthermore, tests on paste and mortar samples with pure fly ash as well as with lignite fly ash cements containing 10, 20 and 30 % by mass of fly ash, produced by combined grinding of cement clinker and fly ash, were performed regarding soundness, compressive strength and durability [15].

The chemical analysis of fly ash samples from different mining areas as given in table 2 was confirmed by this investigation. It was further confirmed that the requirements regarding reactive calcium oxide is not met from fly ash from Lusatian area which can be assigned to class W 1 according to DIN EN 197-1 because the content of reactive calcium oxide is higher than 10 % by mass. The fly ashes of the Rhenish and Central German area can be assigned to calcareous fly ashes W 2 due to high amount of lime of 30 to 37 % by mass and reactive calcium oxide of 26 to 34 % by mass. However, the requirements for compressive strength of mortar samples with ground fly ash as binder (> 10 MPa) could not be met. The requirements regarding expansion of a paste consisting of 30 % by mass of ground fly ash and 70 % by mass of cement are met by all samples.

As expected, mortar samples produced with this lignite fly ash cements show decreasing compressive strengths with increasing amount of lignite fly ash in cement at early ages up to 28 days. The compressive strength of all mortars increased with age. At an age of 28 days mortars with 10 % fly ash in cement show higher compressive strength than the reference mortar with OPC, those with 20 % fly ash after 90 days (see table 3).

Similar results were gained in tests with two different types of OPC and calcareous fly ash from the midth German and Lusatian mining area. Mortars produced by exchange of 20 % by weight of cement by fly ash from midth German area showed similar or higher compressive strength after 28d compared to pure cement mortars. Mortars with exchange of 20 % by weight of cement with fly ash from Lusatian area showed 85 to 100 % of the compressive strength of the pure cement mortars [16]. For these tests the samples were stored at 20 °C as well as at 5 °C.

In 2010, Dyckerhoff AG and MUEG mbH produced a cement with calcareous ash within a joint research project [17]. The fly ash was produced in a midth German power plant and showed a free lime content of 6.5 % by mass. Before use in cement the fly ash was partly slaked in a processing plant. This beneficiation resulted in less than 3 % by mass of free lime and a dry product which could be loaded with air pressure. The chemical composition of the fly ash used for the production of a CEM II B-M (W-LL) is given in table 4.

The cement was used to produce a base plate of 3000 m² at the site of MUEG. In total 660 m³ of concrete with a cement content of 360 kg/m³ with water/binder ration of 0.45 was placed. The concrete was placed as normal concrete and according the construction company no performance deviations compared to normal concrete was observed during concreting. On the base plate a hall was build and the plate is used by container trucks. After two years under service no cracks or damages have been observed [17].

4.5 Hydraulic road binders

The use of lignite fly ash in hydraulic road binders was subject of several research projects [18, 19]. Based on the results of this research projects lignite fly ash was used in hydraulically bound mixtures and hydraulic road binders. Due to the revision of the European standard for hydraulic road binders and hydraulically bound mixtures the German Federal Ministry of Transport, Building and Urban Affairs ordered a project on the suitability of mixtures for hydraulically bound base courses according to European codes for applications in Germany in 2004 [20]. The aim of the research project was to gather the experience in Europe with Hydraulic Road Binders (HRB) and gained with the European standards EN 14227, parts 2 to 4 with slag bound and fly ash bound base course mixtures and to find out whether and under which conditions they can be used for the construction of base courses which fulfil the requirements of the existing national regulations. Within the research project different tests roads built with slag bound and fly ash bound base courses in Germany from 1985 to 1998 had been investigated regarding the bearing capacity by Benkelman beam and Falling Weight Deflectometer. Core samples were tested regarding compressive strength and durability. Aim of the research work was to evaluate whether the requirements in the European standards 14227 part 2 to 4 are sufficient to cover also the experience gained with the German regulations. The results of the investigations revealed that the introduction of the European Standards for slowly hardening slag bound and fly ash bound mixtures in Germany can be generally recommended. It was verified that suitable slag bound and fly ash bound mixtures are sufficiently resistant against frost and have a high long-term durability under the prevailing climatic conditions in Germany, too [21].

4.6 Concrete addition

In the former GDR lignite fly ash was used as a constituent of cement but also as an addition to concrete [9, 22]. The use of lignite fly ash for concrete was based on the former (east) German standard BS 50-0808 [23] for fly ash for concrete, mortars, stabilisation and special masses, which - for the fly ash properties - referred to the former (east) German fly ash standard TGL 190-72/03 “Electric Power Stations: Solid Combustion Residues of Electric stations firing pulverized brown coal; Electric power stations produced dry ash – technical terms of delivery” [24]. With the reunification of Germany in 1989 the standards were replaced by DIN standards. The experience for the use of lignite fly ash for cement and concrete based on the former standards was not taken over into the existing DIN standard for cement and concrete.

According to the definition of fly ash in DIN EN 450 “Fly ash for concrete” [25] fly ash from lignite combustion was excluded from this application covered by the standard although the criteria for siliceous fly ash were met. In Germany, lignite fly ash could only be used as concrete addition if a technical approval from the German building authorities has been granted. In 1990, a technical approval for fly ash from lignite as concrete addition from the Lusatian area was issued by the German building authority which is extended regularly due to continuous use of fly ash from this power plant in the concrete industry [26]. With this technical approval the operator is committed to a quality control system consisting of continuous production control by the producer and a third party control on the fly ash defined to conform to the requirements for fly ash for concrete. The results of the continuous production control are subject to monitoring by a notified certification body.

5. Summary

In German lignite fired power stations about 8.7 million tonnes of fly ash, 1.7 million tonnes of bottom ash and 5.7 million tonnes of flue gas desulphurisation gypsum were produced in 2010. Most of the ashes are used for the refilling and reclamation of depleted opencast lignite mines. Furthermore, they are used as filler in asphalt, in underground mining, for surface restoration, soil beneficiation and as addition to concrete. FGD gypsum is used in the gypsum and cement industry.

The utilisation of the coal combustion products (CCPs) is depending on their chemical, mineralogical and physical properties. These properties are influenced by the power plant technique, the source of coal as well as the type of coal feed. Lignite fly ashes from the main German mining areas show characteristic chemical parameters. Nearly all lignite fly ashes consists of more than 10 % by mass of reactive calcium oxide and may be characterized as calcareous fly ash as defined in DIN EN 197-1.

But the fly ashes do not meet all the requirements of DIN EN 197-1 and therefore do not comply with this standard.

For all specified applications requirements on technical and environmental parameters of national or European regulations or standards have to be considered. For the utilisation in applications where the self-hardening properties of lignite fly ash are required as well as in binder systems with or without cement a constant product quality is prerequisite for utilisation.

Most of the produced ashes from lignite combustion are used for refilling of depleted opencast and underground mines. Many examinations of the use in cement-containing systems have proven that lignite fly ash produced in lignite-fired power plants can be used in many different ways. For the use in hydraulic road binders long term experience have been reported showing good performance after many years of service. The ashes are also regularly used for soil beneficiation in road construction and as concrete addition. A 3000 m³ bottom plate was concreted with a CEM II-B (W-LL) produced in a joint research project with a cement producer.

In principle, lignite fly ash can be used in the building materials and construction industry (cement, concrete, plaster and mortar, road construction) as well as in environmental sectors (retention of pollutants, soil rehabilitation). The selection of fields of application reflects the consideration of economic aspects as well as criteria stemming from the requirements of qualifying examinations, licensing and standardization procedures.

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Table 1 Production and utilisation of CCPs from lignite-fired power plants in Germans in 2010 [2]

	Capacity in [MW_{th}]		68.870	
	Burnt coal in [million tonnes]		150	
	CCP	production	utilisation	
			restoration / reclamation	others
		million tonnes	%	%
	Bottom Ash	1,63	97	3
	Fly Ash	8,47	97	3
	FBC-Ash	0,33	100	
	FGD-Gypsum	4,83	5	95*
	Total	15,26	62	38

* 16 % by mass of this amount on temporary stock

Table 2 Fluctuations of major and minor chemical parameters of lignite fly ash from German power plants in different mining areas [3]

	Mining Area		
Parameter	Rhenish Area	Central German Area	Lausitz Area
	[% by mass]		
SiO_2	20 – 80	18 – 36	32 – 68
Al_2O_3	1 – 15	7 – 19	5 – 14
Fe_2O_3	1,5 – 20	1 – 6	6 – 22
CaO	2 – 45	30 – 52	8 – 23
CaO_{free}	2 – 25	9 – 25	0,1 – 4
MgO	0,5 – 11	2 – 6	2 – 8
K_2O	0,1 – 1,5	0,1 – 0,5	0,5 - 2
Na_2O	0,1 – 2	0,01 – 0,2	0,01 - 0,2
SO_3	1,5 – 15	7 – 15	1 – 6
TiO_2	0,1 – 1	0,5 – 1,3	0,2 – 1
Cl	< 0,2	< 0,1	< 0,02
C	< 2	< 1	< 2
LOI	max 5	max 5	max 5

Table 3 Flow and relative compressive strength of mortars with lignite fly ash cements containing 10, 20 and 30 % by mass of lignite fly ash [13]

Proportion of Lignite Fly Ash in Cement from		Flow	Relative Compressive Strength at			
Rhenish Area	Lausitz Area		2 days	7 days	28 days	90 days
[% by mass]		[mm]	[%]			
0	0	150	100,0	100,0	100,0	100,0
10		150	96,6	95,6	103,5	109,5
20		153	89,9	84,9	93,0	100,6
30		169	87,6	80,5	81,8	89,2
	10	167	98,7	93,1	102,3	110,6
	20	163	94,6	81,9	90,5	102,1
	30	165	81,9	71,4	86,7	95,8

Table 4 Chemical composition and special parameters of fly ash used for production of CEM II B-M (W-LL) 32,5R [17]

Parameter	[% by mass]
SiO_2	28.5
Al_2O_3	12.6
Fe_2O_3	7.2
CaO	35.5
CaO_{free}	< 3
MgO	2.3
K_2O	0.6
Na_2O	< 0.2
SO_3	9.1
CaO_{react}	27
SiO_{2react}	23,1

Fig. 1 Lignite mining areas in Germany [1]

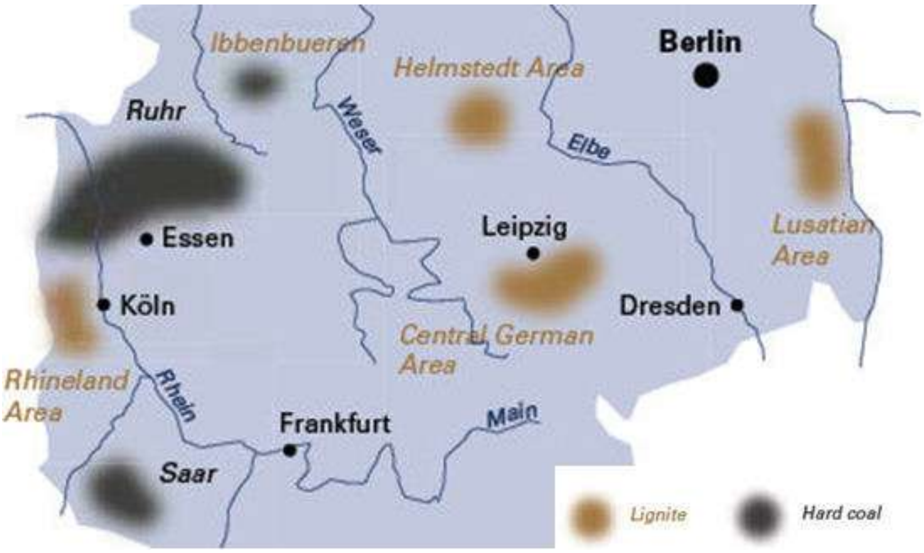

Fig. 2 Combustion process and CCPs in a lignite fired power plants

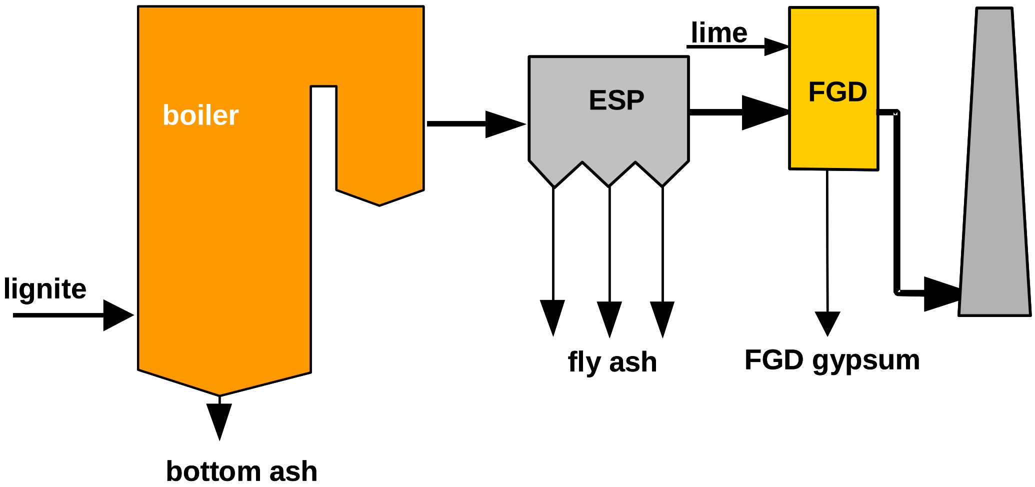

Fig. 3 Base plate with concrete produced with CEM II B-M (W-LL) – MUEG [17]