Solid and fly ash materials ofbrown coal power plants, their characteristics and utilisation

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In coal-fired power plants, a significant amount of residues is produced, depending on the technical parameters of coal separation and firing equipment. A large quantity of solid and fly ash and, in the case of flue gas desulphurisation, REA gypsum and wash-water is produced. The quantity of residues depends primarily on the ash and sulphur content of the fuel.

Coal has a significant role in energy production and represents a considerable quantity in electric energy generation. At the turn of the millenary, about 4 billion tones of black coal and 800 million tones of brown coal and lignite are produced in the world annually. Depending on the ash content of the coals – it varies between 5-8% and 30-35% –, the quantity of solid and fly ash produced by firing is 1.0-1.5 billion tones per year. The quantity of residues of this kind accumulated in the past amounts to 100 billion tones.

As far as the residues of coal-fired power plants are concerned, the annual fuel demand of the power plants of the Rhenish brown coal basin, where the average ash content of lignite is 7% and the average sulphur content is 0.2-0.8%, is 1 Mt referred to a power plant capacity of 100 MW. 60-70 kt solid + fly ash and, in the case of flue gas desulphurisation, 12-15 kt of gypsum is produced annually, referred to a capacity of 100 MW. In the East German areas, after the reconstruction of power plants, 30-50 kt of fly ash and, because of the higher sulphur content, 25-30 kt of gypsum and 4-5000 m^3 of wash-water is produced annually, referred to a capacity of 100 MW.

The composition of Hungarian lignite is significantly different to that of Rhenish brown coal. The ash content and combustible sulphur content of domestic lignite is considerably higher. The ash content of lignite varies between 15 and 25%, the average is 20%. In Visonta, 160-200 tones of solid + fly ash is produced annually, referred to a power plant capacity of 100 MW. With the flue gas desulphuriser installed recently, one can expect a gypsum quantity of 40-60 kt/year, referred to a capacity of 100 MW.

Chemical composition and grain size distribution of solid and fly ashes

The chemical/mineral composition and grain distribution of solid and fly ashes is important for the utilisation possibilities and, in the case of deposition, these parameters influence solidity, water permeability and infiltration coefficient. The solid and fly ash from coal-fired boilers is dust-like, practically without any solidity. The solid ash is coarser and the fly ash is finer, with a very loose structure. For the residues of the Visonta Power Plant, the density of solid ash is 1650-1750 kg/m^3, that of fly ash is 2000-2050 kg/m^3 and that of solid + fly ash mixture is 1900-2000 kg/m^3. During hydraulic delivery, materials are disintegrated and the density of the disintegrated solid + fly ash mixture is 2150-2300 kg/m^3, nearly identical with that of sand.

According to measurement data, it is typical of fluid-bed and dust coal-fired boilers that most of the SiO_2 content leaves in the solid ash – the SiO_2 content of solid ash is 60-90% – and only a small part remains in the fly ash – the SiO_2 content of fly ash is 20-50%. At the same time, most of the CaO +MgO content appears in the fly ash. Similarly, most of the Fe_2O_3 + Al_2O_3 content can be found in the fly ash. The proportion of the individual elements in the different materials varies within a relatively wide range. The upper limit may be 10-15 times the lower one.

Table 1 shows the characteristic data of the chemical parameters of the solid + fly ashes of different mining areas. The average values of the SiO_2 content are nearly identical. For brown coal and lignite fired power plants, the SiO_2 content is between 45 and 60%. The CaO + MgO content determining the hydraulic (setting) properties of residues significantly differs for the individual materials. It is 10-12% in Visonta, 20-25% in Inota and 20-30% in the Rhenish area. However, the Rhenish area shows the lowest Fe_2O_3 + Al_2O_3 content (12-18%). It has a high value (25-30%) in Visonta and a medium value (20-25%) in Inota. These proportions are largely responsible for the fact that the material of the deposit of Visonta does not solidify at all after hydraulic delivery and drying, while in the Rhenish area a strength of 5-20 MPa can be measured.

We displayed the data of chemical composition in Rankin's diagram (Figure 1), together with the parameters of other materials. The situation of the areas characterising the composition of the individual

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materials shows that the composition of the brown coal ash of Germany and Inota (Balinka) is significantly different to that of the deposit material of Visonta.

Tab.1 Characteristic data of the chemical composition of solid and fly ashes from different mining areas (in mass percent without firing loss)

			SiO_2	CaO	MgO	CaO + MgO	Fe_2O_3	Al_2O_3	Fe_2O_3 + Al_2O_3
1.	Black coal ash(fluid-bed firingin full mass%)		33-48	8-18	1-2	9-20	5-10	13-22	18-32
2	Black coal ash (fluid-bed firingin dry mass%) Germany		40-55	1-8	0.4-4.8	2-13	4-17	23-35	27-52
3	Brown coal ash (in dry mass%) Germany		20-70	8-40	0.5-7	9-47	1.5-20	1-15	2.5-35
4	Rhenish brown coal (coal dust firing)		45-58			20-28			12-18
5	Visonta-Bükkábrány seams		41-49			10-12			28
6	Visonta Power Plant (coal dust firing)	samples	45-60	5-10	1.5-25	6.5-12.5	8-14	12-18	20-32
		average	53			10			26
7	Ash lagoon of Inota	samples	45-53	18-26	2-3	20-29	7-9	14-17	21-26
		average	50	21±3	2.6±0.6	23-24	7.5±0.7	15±1.0	22-23
8	Inota, boilers 3 and 5, filter fly ash		51-53	18-20	2-2.2	20-22	7-7.5	16-16.5	

The grain size composition and distribution of solid and fly ashes is important for the application possibilities and, in the case of deposition, these parameters influence solidity and water permeability. The usual grain size functions characterise the material composition – the "steeper" the function, the more homogenous the material and the lesser the need for classification and separation by grain size.

Figure 2 shows the grain size composition and the grain distribution functions for materials of different areas. About 80 mass% of the Rhenish fly ashes has a grain size less than 50 μm, so it has favourable hydraulic properties. This proportion is 70% for the fly ashes of Inota. In Visonta, however, only 20-30 mass% of the fly ash falls in the range under 0.05 mm. Apart from the low CaO +MgO content, the latter fact is the reason why the material of the deposit of Visonta does not solidify.

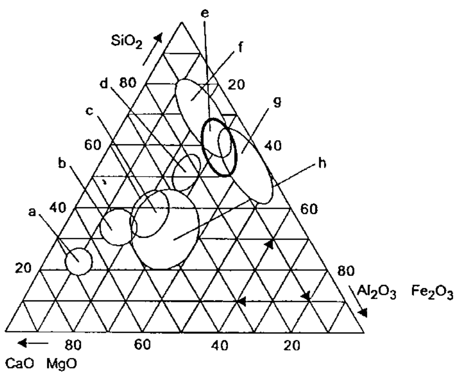

Fig.1 Analysis results for the materials of different industrial areas (brown coal, cement, waste, black coal and pozzolana) in Rankin's diagram. a- Portland cement; b – high-furnace cement; c – refuse burning plant fly ash dust; d - Inota Power Plant solid + fly ash; e - Visonta Power Plant solid + fly ash; f – natural pozzolanas; g – black coal fly ash; h - brown coal fly ash (Germany).

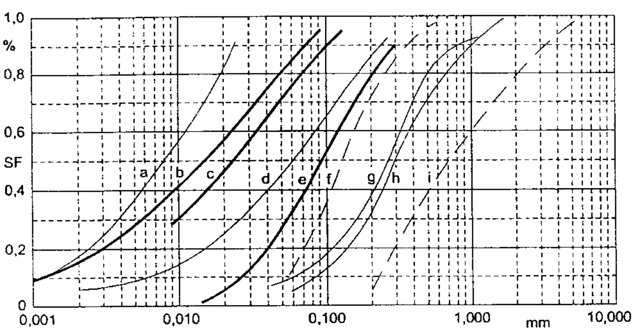

Fig.2 Grain size distribution functions for solid and fly ashes of different areas. a - VEAG black coal; b – Rhenish fly ash (average); c –fly ash of Inota; d - fly ash of Braunschweig; e – fly ash of Visonta (average); f – German solid ahs (lower limit); g – solid ash of Inota; h – Rhenish solid ash (average); i – German solid ash upper limit).

The grain size of ashes is two orders of magnitude larger than that of materials with favourable hydraulic properties (e.g. cement). The relatively wide – thousandfold – grain size range explains the fact that power plant residues have very different properties.

Solidity characteristics of solid + fly ash materials and permeability of deposits

After deposition, the dry material of Visonta has a very loose structure and porous property, it is water-permeable and its strength is practically zero. According to the data of foreign literature, the internal friction angle of mixed (solid + fly) ash varies between 27º and 36º. The internal friction angle of the solid/fly ashes of Visonta is within this range.

According to laboratory measurements, the infiltration coefficient of dry solid + fly ashes is $k = 10^{-4} - 10^{-5}$ m/s and the value after self-weight compaction is $k = 7 \cdot 10^{-5}$ m/s, in some places 10^{-6} m/s. According to similar measurements, the infiltration coefficient of dumped dead material from surface mining overburden stripping is around $k = 4 \cdot 10^{-5}$ m/s.

The hydraulic and solidifying properties of the individual materials – setting agents – are primarily determined by the chemical and grain size composition. In this regard, the chemical composition is determined by the hydraulic, silicate and aluminate modulus as well as the so-called basicity. During the examination of solid and fly ashes, these parameters are compared with the usual value limits given for Portland cements. Highlighting only the hydraulic modulus:

$$HM = \frac{CaO(\%)}{SiO_2(\%) + Fe_2O_3(\%) + Al_2O_3(\%)}$$

we obtain that this ratio is $HM_{average} = 0.04 – 0.07$ for the solid + fly ash samples from Visonta and $HM_{average} = 0.40 – 0.60$ for the Rhenish brown coal solid + fly ashes. The usual value given for Portland cements is $HM_{average} = 1.7\text{-}2.3$.

The HM values of the ash of Visonta are one order of magnitude lower than the characteristics of Rhenish fly ashes and two orders of magnitude lower than the hydraulic modulus of cement. The $CaO:SiO_2$ ratio of the solid + fly ashes of Visonta usually does not reach this limit (0.1:1.0), above which the hydraulic property of brown coal comes to force. For Rhenish fly ashes, this ratio is 0.1-0.5 .

Another characteristic of hydraulic properties is the grain size of setting materials. The upper limit of the required grain size is 0.03-0.05 mm. The bulk of Rhenish fly ashes (Figure 2) is under this grain size range, while in the ash lagoon samples of Visonta only 5-25 mass% falls in the grain range finer than 50 µm.

Our solidification experiments with the solid + fly ashes of Visonta showed that these materials show no solidifying properties (similarly to the phenomena experienced at the deposit).

The permeability of the deposits and the strength factors can be determined simultaneously with the solidity tests. The extent of dissolution of materials in the deposit (chlorides, sulphates, toxic materials and heavy metals) is significantly influenced by the permeability of the solidified complex. The results of permeability measurements show that the increase of the chloride and sulphate content decreases the value of the k factor.

The extent of decrease depends on the CaO + MgO content of the deposit material. With a high CaO content, the addition of REA water (chloride and sulphate) may decrease the k value by three orders of magnitude. The addition of REA gypsum, however, increases permeability and decreases solidity.

Figure shows the change of the k factor plotted against the water content of the material transported to the deposit, according to the data of the Rhenish area. In the chemical conversion process of solid + fly ash, the stoichiometrically required water quantity (25-35%) results in a nearly maximum solidity with a minimum permeability. At he same time, adding water in this quantity does not cause any unnecessary dissolution and wash-out of hazardous material.

Figure 4 shows the change of the infiltration coefficient plotted against the crushing strength of the dump material, also according to the Rhenish data.

During depositing the residues, the dissolution from the deposit material and the environmental hazard of the dissolved material may be a serious hazard both in the short and the long run. The dissolved materials are primarily chlorides and sulphates and secondarily heavy metals and toxic materials. The maximum dissolution concentration of chlorides and sulphates may reach 4,000-10,000 mg/l. The extent of dissolution decreases simultaneously with the setting of the deposit material and the conversion and phase transition of the minerals. According to measurements in Germany, the chloride concentration decreases to 100-500 mg/l and the sulphate concentration to 200-400 mg/l after a setting time of 28-30 days. After a setting time of 180 days, the dissolution concentration is 40-200 mg/l for chlorides and 100-200 mg/l for sulphates.

The German regulations permit a chloride (Cl^-) concentration of 200 mg/l and a sulphate ($SO_4^{\ 2+}$) concentration of 240 mg/l for drinking water. German publications state that the heavy metal (ion) dissolution from the fly ash or the deposit always remains under the permissible limit value.

The radiating material content of coals and solid/fly ashes may be relevant concerning environmental effects. The examination of the uranium content of domestic coals showed that the Cretaceous stone coal of Ajka has the highest uranium content in Hungary. The uranium content of the lower seams in Ajka may reach 0.1%, i.e. 1000 g/t, with an average value of a few 100 g/t.

The uranium content of the Transdanubian Eocene coals is relatively high – usually a few 10 g/t, with a maximum value of 300-400 g/t.

The uranium content of the Lias coals of the Mecsek mountains is relatively low, around 10 g/t. The uranium content of the Miocene brown coals of North Hungary and the lignites of the Mátra mountains and Bükkalja – and of course their ash – is a few g/t.

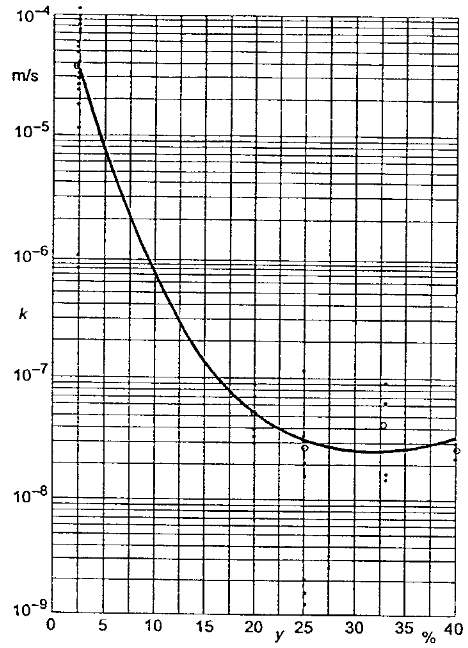

Fig.3 Change of the infiltration coefficient plotted against the water content of deposited fly ash (during dry and wet deposition of Rhenish brown coal fly ash).

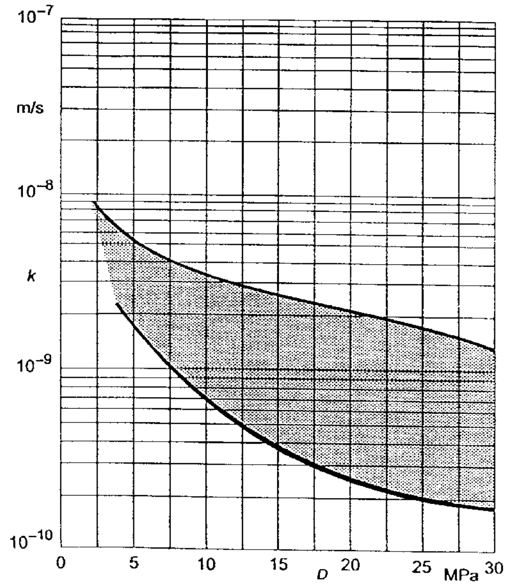

Fig.4 Change of the infiltration coefficient plotted against the crushing strength (Rhenish brown coal area)

Utilisation of solid and fly ashes

At power plants operating with flue gas desulphurisation, solid and fly ash is usually treated together with REA water. The filling of surface mining cavities – mainly to reduce the environmental effects – is done in a special so-called cassette form. Where there is – or there was – no flue gas desulphurisation, fly ash is available in larger quantities for other application purposes.

The utilisation of fly ash for other purposes is usually restricted by the following factors:

- The fly ash quality is not uniform (permanent) as it depends on the quality of the coal seam, the surface mining technology and the firing parameters.
- The produced or previously accumulated fly ash is immobile an has a relatively large quantity.
- The preparation of fly ash is not profitable because of the price of substitutes (traditional construction materials).

Intensive research work is going on concerning the utilisation of solid/fly ashes. In Germany alone, 15 research programs are in progress, connected with pilot- and large-scale experiments.

Utilisation possibilities and research directions:

- production and extraction of industrial minerals (e.g. zeolite, wollastonite and aluminium oxides);
- utilisation as cement and concrete additive;
- utilisation as construction material (production of light and heavy building elements, utilisation as setting material in the case of a high CaO+MgO content, substitution for sand in the case of a high SiO_2 content);
- utilisation of hydraulic fly ashes (brown coal/lignite fly ash) for constructing roads, embankments and dams;
- stowing of surface mining and underground cavities;
- soil improvement and recultivation.

Analysing the environmental issues of utilisation, the chemical test of different materials (cement, refuse burning plant and brown coal fly ash) showed that the quantity of Na, K, Co, Fe and Al in the tested dusts is essentially identical. The concentration of Cr, Zn, Pb, Cu, Co, Cl and Ni is significantly lower in brown coal (lignite) fly ashes than in other materials. The latter three elements do not occur at all in Rhenish fly ashes.

The results of the research work showed that the heavy metals (trace elements) found in brown coal (lignite) fly ashes do not cause any environmental hazard during deposition and the dissolution remains under the permissible limit values.

Literature

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