COAL COMBUSTION PRODUCTS IN EUROPE
- PRODUCTION, UTILISATION, STANDARDISATION -

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ABSTRACT: In Europe, coal combustion products (CCPs) are mainly utilised as a replacement for natural materials in the building material industry, in civil engineering, in road construction, for construction work in underground coal mining as well as for recultivation and restoration purposes in open cast mines. The majority of the CCPs is produced to meet certain requirements of standards or other specifications with respect to utilisation in certain areas.

The utilisation of fly ash in Europe is being influenced by legislation (Waste Directive, REACH Regulation). The use of CCPs has several environmental and technical benefits. It has developed by the years and is based on requirements of standards or other specifications which are subject to regular revision by CEN or national authorities. The paper focuses the changes in the revised Waste Directive, the requirements according to the REACH regulation as well as the revision of European standards EN 450 fly ash for concrete.

1 INTRODUCTION

Coal combustion products (CCPs) are produced with the production of electricity in coal-fired power plants. Coal ash is a synonym for the combustion residues boiler slag, bottom ash and especially fly ash from different types of boilers. These ashes form the major part of all coal combustion products (CCPs) which consist also of desulphurisation products like spray dry absorption product and FGD gypsum.

In 2007, about 61 million tonnes of CCPs were produced in Europe (EU15). The production in all the European member states is estimated to be about 100 million tonnes. The utilisation of CCPs is well established in some European countries, based on long term experience and technical as well as environmental benefits. The CCPs are mainly utilised in the building material industry, in civil engineering, in road construction, for construction work in underground coal mining as well as for recultivation and restoration purposes in open cast mines. The majority of the CCPs is produced to meet certain requirements of standards or other specifications with respect to utilisation in specific areas.

The utilisation of CCPs in Europe is being influenced by environmental legislation and by the liberalization of the electricity market. At present, the most important discussion focuses on the definition of CCPs. With the revision of the Waste Directive also “by-products” and “end-of-waste” will be defined. By-products will from the beginning not be subject to waste legislation; end-of-waste materials are resulting from the recovery of waste. For non-waste materials the REACH regulation has to be considered and substances which are not waste are subject to REACH.

The use of CCPs has several environmental and technical benefits. It has developed by the years and is based on requirements of standards or other specifications which are subject to regular revision by CEN or national authorities. At present, the European standards EN 450-1 and EN 450-2 fly ash for concrete are under revision. Also under revision are the new revised
standards for hydraulic road binders, for hydraulic mixtures, for aggregates and for lightweight aggregates.

2 PRODUCTION AND UTILISATION OF CCPS

2.1 Production

The ECOBA statistics on production and utilisation of CCPs [ECO07] reflect the typical combustion products such as fly ash (FA), bottom ash (BA), boiler slag (BS) and fluidized bed combustion (FBC) ashes as well as the products from dry or wet flue gas desulphurisation, especially spray dry absorption (SDA) product and flue gas desulphurisation (FGD) gypsum.

In figure 2.1 the development of CCP production in EU 15 member states from 1993 to 2007 is shown. The total amount decreased from 57 million tonnes in 1993 to 55 million tonnes in 1999 and rose again to 64 million tonnes in 2005 due to higher production of electricity and heat by coal combustion.

In 2007, the amount of CCPs produced in European (EU 15) power plants totalled 61 million tonnes, about 3 million tonnes less compared to 2005. This reduction was caused by smaller production by coal combustion in some countries due to higher production by hydro power or the installation of de-NOx and de-SOx measures. In 2007, all combustion residues amounted to about 82 % and the FGD residues to about 18 % by mass.

![Fig. 2.1. Development of the CCP production in Europe (EU 15) from 1993 to 2007. [ECO07].](image)

In contrast to the expectations based on the overall discussion about CO₂ reduction an increase of the total amount of CCPs is expected due to retrofitting of existing coal fired power plants with FGD based on requirements of the Large Combustion Plants Directive (LCPD), the construction of new coal fired power plants in some countries as well as the increased use of imported coal with higher ash content.
2.2 Utilisation of CCPs

The CCPs are mainly utilised in the building material industry, in civil engineering, in road construction, for construction work in underground coal mining as well as for recultivation and restoration purposes in open cast mines. In 2007, about 53\% of the total CCPs are used in the construction industry, in civil engineering and as construction materials in underground mining and about 36\% for restoration of open cast mines, quarries and pits. About 2.5\% were temporarily stockpiled for future utilisation and about 8\% were disposed off. The rates of utilisation, temporary stockpile and disposal for the single CCPs are given in figure 2.2.

![Utilisation, temporary stockpile and disposal of CCPs in Europe (EU 15) in 2007](ECO07)

The fields of utilisation of specific CCPs in 2007 in EU 15 are described below. Figures regarding the utilisation of specific CCPs in 2007 in EU 15 countries are given in Annex 1.

Fly ash: About 20 million tonnes of fly ash were utilised in the construction industry and for production purposes in underground mining. Most of the fly ash produced in 2007 was used as concrete addition, in road construction and as raw material for cement clinker production. Fly ash was also utilised in blended cements, in concrete blocks and for infill (that means filling of voids, mine shafts and subsurface mine workings; see figure A1 in Annex 1).

Bottom ash: Out of a total of 2.5 million tonnes of bottom ash used in the construction industry about 45\% was used as fine aggregate in concrete blocks and in concrete, about 37\% in road construction and filling applications and about 13\% in cement production (see figure A2 in Annex 1).

Boiler slag: About 66\% of the boiler slag produced was used as blasting grid, about 10\% for grouting and in drainage layers and about 14\% was used as aggregate in concrete (see figure A3 in Annex 1).

Fluidized Bed Combustion (FBC) ash: Out of a total of 0.9 million tonnes 0.2 million tonnes were mainly used for engineering filling applications (40\%), for infill (30\%) and subgrade stabilisation (about 14\%; see figure A4 in Annex 1). It has to be noted that the total amount of FBC-ash in EU 15 countries is small compared to the amount produced at least in Poland and the Czech Republic.
Spray Dry Absorption (SDA) product: About 0.3 million tonnes of the total SDA product was utilised in the construction industry and in underground mining (63 %), for plant nutrition (8 %) and as a sorbent in wet FGD (29 %; see figure A5 in Annex 1). It has to be noted that also the total amount of SDA-product in EU 15 countries is small compared to the amount produced at least in Poland and the Czech Republic.

FGD Gypsum: Out of the total of 8.8 million tonnes about 62 % was used for the production of plaster boards. Other applications include the production of gypsum blocks, projection plasters and self levelling floor screeds (31 %). In the cement industry FGD gypsum is used as set retarder (7 %; see figure A6 in Annex 1).

3 LEGAL ISSUES

3.1 Revision of the Waste Directive

According to the European Waste Framework Directive from 1996 CCPs have legally to be considered as waste. Since the early 90ties of the last century discussions took place on the question if in certain cases a by-product from industrial processes is covered by the definition, i.e. has to be considered as waste or if waste properties could cease at a specific stage of the managing process. A typical example is FGD gypsum, which is on one hand a residue of a pollution abatement process and on the other hand was produced from scrubber sludge by an oxidation, cleaning and drying procedure aiming at a material, which meets technical specifications of the users (gypsum and cement industry). After several years of discussion it is now generally accepted by the authorities that FGD gypsum has ceased the waste properties after the processing in the power plant. The case is not clear for fly ash as it is argued that no processing takes place in the power plant and that the recovery operation is the final use of the material.

Within the revision of the Waste Directive the discussion on the legal definition of by-products and end-of-waste status was restarted at the European institutions and ended with the publication of a revised Waste Directive including definitions for “by-products” and “end-of-waste” status. The Directive entered into force on 12 December 2008 [WDR08]. Member States must adopt the measures to comply with the Directive within 24 months, i.e. by 12 December 2010. By-products according article 5 will from the beginning be not subject to waste legislation, end-of-waste materials are resulting from a recovery process. For both types of materials specific criteria must be met. For “by-products” according to article 5 of the Directive these criteria are:

(a) further use of the substance or object is certain;
(b) the substance or object can be used directly without any further processing other than normal industrial practice;
(c) the substance or object is produced as an integral part of a production process; and
(d) further use is lawful, i.e. the substance or object fulfils all relevant product, environmental and health protection requirements for the specific use and will not lead to overall adverse environmental or human health impacts.

Due to these criteria the CCP producers are of the opinion that CCPs are by-products as the production process is fully controlled to meet the requirements of emission regulations as well as of standards and specifications and as the product is offered to the existing construction
material markets [EUR06]. However, with the implementation of the revised Waste Directive in the EU member states additional criteria may be defined on national level [WDR08].

3.2 REACH Regulation

On 1st June 2007, the REACH-Regulation (Registration, Evaluation, Authorisation and Restriction of Chemicals) entered into force [REA06]. By this, also each producer or importer of coal combustion products (CCPs) has to register the marketed or imported substances at the European Chemicals Agency (ECHA) situated in Helsinki. Since 1 June 2008, CCPs which are not registered can not be produced and placed on the market.

Exemption: For CCPs which have been pre-registered by 1 December 2008 the deadline for registration is extended by 1 December 2010 (production volume > 1000 tonnes).

The producers/importers of CCPs have pre-registered FGD-gypsum, spray dry absorption product (SDA-product), fly ash ((FA) from hard coal/lignite), bottom ash (BA), boiler slag (BS) and fluidised bed combustion ash (FBC-ash) as well as cenospheres (CE) with the CAS- and EC-numbers of the specific substances.

One option to organise oneself and to start the substantial work for the joint registration in a SIEF is the formation of consortia. By now, consortia for calcium sulphate, ashes (fly ash (including cenospheres), bottom ash and boiler slag) and for FBC-ash have been formed.

The producers/importers of substances with the same EC-numbers form so-called pre-SIEFs (Substance Information Exchange Forum). At present, the pre-registered parties discuss the sameness of substances (SIP: Substance-Identity-Profile). Sameness is the pre-condition for the formation of a SIEF and the preparation of a joint registration dossier with chemical, physical, toxicological and ecotoxicological data. For the registration of one substance only one registration dossier with the specific registration data can be filed. A so-called „Lead Registrant“ will submit a substance specific joint registration dossier to the ECHA. The lead registrant will provide each interested member of the consortium with the joint submission name and token which it has received by the ECHA via REACH-IT after formal check of the joint dossier. The information is essential for filing the producer-/importer- specific dossiers to ECHA.

4 REVISION OF EUROPEAN STANDARDS

4.1 Revision of EN 450-1 and EN 450-2

The application as concrete addition constitutes the highest added value for fly ash. By this, the European Standard EN 450 "Fly Ash for Concrete" is particularly important for the marketing of fly ash. The standard was first published in 1994 [EN100] and the revised standards EN 450-1 und EN 450-2 entered force on January 1, 2007 [EN107; EN205]. EN 450-1 deals with definitions, specifications and conformity criteria for siliceous fly ash, which is produced by burning of pulverized coal, with or without co-combustion materials, and collected in a dry state, or which is processed by e.g. classification, selection, sieving, drying, blending, grinding or carbon reduction or by a combination of these processes. This is because in some countries fly ash has been processed according to national regulations for
years or, in some cases, decades. EN 450-2 deals with the conformity evaluation of fly ash for concrete produced in power plants and in processing plants. Most important is the documentation of procedures for the production control in a works quality manual.

In EN 450-1 requirements regarding homogeneity, soundness and effectiveness are stipulated. The chemical requirements refer to e.g. the loss on ignition, sulphur-, chloride- and free-lime-content. If fly ash is produced with co-combustion the content of reactive SiO$_2$, the total oxide content of SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, the alkali-, MgO- and phosphate-content have to be tested. The physical parameters stipulate requirements on the fineness, variation of fineness and density, on soundness and activity index. In addition to these requirements, fly ash from co-combustion has to meet the requirement of initial setting time and fly ash of category S for water demand.

Within the ongoing revision of the standards all parameters will be critically reviewed and the amount of co-combustion materials will be increased to incorporate the experience gained with European Technical Approvals (ETA). All proposed changes will be documented in a respective background report which will be published as CEN report after final review [BGR09].

Within the revision of part 1 of the standard the definition of fly ash and the amount of co-combustion materials and the requirements for chemical parameters (LOI, free lime, reactive SiO$_2$, and phosphate) will be modified. In part 2 mostly editorial changes referring the definition of the management representative, the methodology for the calculation of the acceptability constant $k_A$ and the requirement for the evaluation of the accuracy of the autocontrol testing will be adapted.

In the definition of fly ash the criterion for reactive silicon dioxide will be deleted due to formal reasons. The reactive silicon dioxide content will only be detected in the initial type testing for new fly ash as all test results showed that the requirement is always met. The amount of co-combustion materials will be increased from 20 to 40 % and the proportion of ash derived from co-combustion material from 10 to 30 % to include the experiences gathered with European Technical Approvals over the last years. For green wood, e.g. not recycled wood, the maximum percentage of co-combustion material can be increased to 50 % by mass.

Although the increased shares seem to be high it has to be noted that the rates can not be reached for all co-combustion materials. Only for green wood the co-combustion will be limited on fuel base. This was covered with the experiences from European Technical Approvals (ETAs). For other types of solid bio fuels and co-combustion materials the shares will be limited by chemical requirements (see table 4.1).
Table 4.1. Limiting parameters for co-combustion materials according table 1 of EN 450-1 [BGR09]

<table>
<thead>
<tr>
<th>Type</th>
<th>Limited by</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Solid Bio Fuels complying with CEN/TS 14588 including animal husbandry residues</td>
<td></td>
</tr>
<tr>
<td>green wood 1</td>
<td>Co-combustion fuel based</td>
</tr>
<tr>
<td>green wood 2</td>
<td>Co-combustion fuel based</td>
</tr>
<tr>
<td>green wood 3</td>
<td>Co-combustion fuel based</td>
</tr>
<tr>
<td>bark wood</td>
<td>reactive CaO</td>
</tr>
<tr>
<td>Cacao shells</td>
<td>Na₂O equivalent (K)</td>
</tr>
<tr>
<td>palm kernels</td>
<td>total P₂O₅</td>
</tr>
<tr>
<td>poultry dung</td>
<td>reactive CaO</td>
</tr>
<tr>
<td>2 Animal meal (meat and bone meal)</td>
<td></td>
</tr>
<tr>
<td>meat &amp; bone meal</td>
<td>total P₂O₅</td>
</tr>
<tr>
<td>3 Municipal sewage sludge</td>
<td></td>
</tr>
<tr>
<td>municipal sewage sludge</td>
<td>total P₂O₅</td>
</tr>
<tr>
<td>4 Paper sludge</td>
<td></td>
</tr>
<tr>
<td>Paper sludge</td>
<td>CaO</td>
</tr>
<tr>
<td>Petroleum coke</td>
<td></td>
</tr>
<tr>
<td>Petroleum Cokes</td>
<td>.*</td>
</tr>
<tr>
<td>6 Virtually ash free liquid and gaseous fuels</td>
<td></td>
</tr>
<tr>
<td>Industrial HC liquid</td>
<td>Co-combustion fuel based</td>
</tr>
</tbody>
</table>

* environmental regulations

For other types of solid bio fuels and co-combustion materials the shares will be limited by chemical requirements. By this, it is guaranteed that the fly ash quality will not change.

The modifications for chemical parameters are described as follows:

- the lower limits of the three classes of LOI will be deleted as the statistical assessment has to be evaluated by variables. This system is made for normal distributed data sets and acceptability constant together with producer and consumers risk. The system was established by D.B. Owen in 1962, but for one sided evaluations only.
- the minimum free lime content above which soundness has to be tested will be increased from 1 % by mass to 1.5 % by mass. Further, the maximum amount of free lime of 2.5 % by mass will be deleted. The changes are based on a data compilation in European countries.
- the determination of the content of soluble phosphate (P₂O₅), determined in accordance with the method described in Annex C, will only be determined in initial type testing. For the continuous proof the content of total phosphate (P₂O₅) will be determined in accordance with EN 196-2 and shall not be greater than 5.0 % by mass. The modification is based on a data compilation in European countries (see figure 4.1).
Fig. 4.1. Relation between the content of total phosphate and soluble phosphate

The modifications for tests methods are described as follows:

- the new standard EN 196-2 allows to use other methods than the classical wet chemical methods, namely X-Ray fluorescence analysis (XRF-EN196-2.2). By this, the use of the XRF-analysis need no longer to be classified as alternative method with the proof of equivalent results but will be used directly as reference method.
- the fineness of fly ash shall be expressed as the mass proportion in percent of the ash retained when sieved on a 0.045 mm mesh sieve. The sieving can be determined in accordance with EN 451-2 with wet sieving or by airjet sieving according EN 196-6.

4.2 Revision of EN 13282 Hydraulic Road Binders

The revision of the European standard for hydraulic road binders ENV 13282 [EN300] resulted in the preparation of three parts. pr EN 13282 Part 1 is dealing with rapid hardening hydraulic road binders [EN309]. These are cement based binders which follow the requirements as already known from ENV 13282. pr EN 13282 Part 2 is dealing with normal hardening hydraulic road binders [EN409]. These binders have lower cement contents, the compressive strength has to be tested after 56 days (part 1 at 28 days). In addition to the main constituents in part 1 also a specific Fluidized Bed Combustion (FBC)-ash and a specific Basic Oxygen Furnace (BOF)-slag with defined chemical and physical parameters can be used. Furthermore, a slaking procedure was implemented to guarantee that also lime rich mixtures can be evaluated in the laboratory. Part 3 of the standard series will deal with the conformity evaluation.

4.3 Revision of EN14227 Hydraulically Bound Mixtures

In June 2009, the revision of the EN 14227-series for hydraulically bound mixtures was decided. The standards under revision cover cement bound (part 1), slag bound (part 2), fly ash bound (part 3) and hydraulic road binder bound (part 5) mixtures. The revision of these standards focuses the structure and the grading of the mixtures. Part 4 deals with fly ash for fly ash bound mixtures and gives the definition and requirements for siliceous and calcareous fly ash.
4.4 Revision of aggregate standards

The European Standards EN 13055- Part 1: “Lightweight aggregates for concrete, mortar and grout” and EN 13055-2 Lightweight aggregates - Part 2: “Lightweight aggregates for bituminous mixtures and surface treatments and for unbound and bound applications” will be merged to one standard according the decision of TC 154. The new standard EN 13055 will be entitled “Lightweight aggregates for building, civil engineering and other applications”.

The standard is relevant for CE marking as lightweight aggregates for the intended use in “Concrete, mortar and grout”, “Bituminous mixtures and surface treatments” and now also in “Unbound and hydraulically bound applications”.

Most of the proposed changes are editorial. Ongoing discussion refer to the scope of the standard, the definitions considering also other aggregate standards (i.e. EN 12620, EN 13242), the test methods described in Annexes and the annex of factory production control and minimum test frequencies. The standard will serve as a first example for the implementation of ER 3 “Dangerous substances”.

5 CONCLUSIONS

In Europe (EU 15) about 61 million tonnes of Coal Combustion Products (CCPs) were produced in 2007. The annual production in EU 27 is estimated to amount to about 100 million tonnes. The CCPs include combustion residues such as boiler slag, bottom ash and fly ash from different types of boilers as well as desulphurisation products like spray dry absorption product and FGD gypsum.

The utilisation is becoming more and more restricted by environmental regulations. A European Technical Committee is working on horizontal standardised assessment methods for the release of dangerous substances from construction materials. In addition, the legal definition of CCPs as waste causes hurdles, which are unnecessarily impeding the utilisation markets, which have been developed in the last decades. With the revision of the Waste Directive a definition of by-products will be introduced for materials which are from the beginning not a waste. Materials may also leave the waste regime after a recovery operation and meeting of waste stream specific end-of-waste criteria. For coal ash as aggregate these may be based on leaching limits.

Materials not being waste are subject to REACH and have to be registered before being placed on the market from 1st June 2008. For CCPs, a special regulation could be used since they are already registered in the European Inventory of the Existing Commercial Chemical Substances (EINECS). By this, the deadline for registration is extended to 1st December 2010 if the producer pre-registered by 1st December 2008! The pre-registered parties have to agree on the sameness of their substances. At present, consortia for the joint registration of calcium sulphate and ash (from dry and wet bottom boilers as well as from FBC-boilers) are formed. Further consortia will be installed to register other types of ashes and other CCPs.

The use of CCPs contributes to sustainability and has several environmental and technical benefits. It has developed by the years and is mostly based on requirements of standards or other specifications which are subject to regular revision by CEN or national authorities. At present, the European standards EN 450-1 and EN 450-2 are under revision. Within the ongoing revision of the standards all parameters are subject to critical review. Proposed
changes will be documented in a respective background report. Furthermore, the European standard for hydraulic road binders with basic definitions which includes FBC ash as a main constituent will be published in an updated version with three parts.

REFERENCES


[EN100] EN 450: Fly Ash for Concrete – Definitions, requirements and quality control, 1994


[EN300] ENV 13282 Hydraulic road binders – Composition, specifications and conformity criteria, 2000


Annex 1

Figure A1:

Figure A2:

Figure A3:
Utilisation of Boiler Slag in the Construction Industry and as Blasting Grid in Europe (EU 15) in 2007. Total utilisation 1.3 million tonnes.

Figure A4:
Utilisation of FBC Ash in the Construction Industry and Underground Mining in Europe (EU 15) in 2007. Total utilisation 0.2 million tonnes.

Figure A5:
Utilisation of SDA-Product in the Construction Industry and Underground Mining in Europe (EU 15) in 2007. Total utilisation 0.3 million tonnes.

Figure A6: