Coal Combustion Products in the United Kingdom and the Potential of Stockpile Ash

Robert A Carroll¹

¹United Kingdom Quality Ash Association, Maple House, Kingswood Business Park, Holyhead Road, Albrighton, Wolverhampton, WV7 3AU, United Kingdom.

CONFERENCE: 2015 World of Coal Ash – (www.worldofcoalash.org)

KEYWORDS: Fly ash, pulverised fuel ash, PFA, stockpile ash, coal fly ash, EN 450, type II addition, concrete, furnace bottom ash, FBA, coal combustion products, CCP

ABSTRACT

Within the United Kingdom fly ash and furnace bottom ash (FBA) are used in the manufacture of a wide range of construction products. Major uses of these valuable by-products are reviewed in this paper.

Sustainable construction products are being sought by specifiers and customers within the United Kingdom. This well established trend is driven by market demand and government initiatives. Fly ash and FBA give proven technical benefits and are included in numerous product standards. The use of these by-products also improves the sustainability of construction materials. A notable example is fly ash produced to European Standard EN 450 and used as a pozzolan in concrete. This by-product is now obtained directly from a number of coal-fired power stations using various processing methods.

Demand for EN 450 fly ash is likely to remain strong or indeed increase in the medium to long term and possible limitations to the supply are discussed. Large amounts of fly ash are held in ash fields and lagoons throughout the United Kingdom. This stockpile ash could provide a complementary supply of fly ash, but requires appropriate processing in order to meet the specification of EN 450 and be an effective pozzolan for concrete. A scheme to assess the potential of stockpile ash is outlined which includes a research project to investigate methods of recovery.

1 Introduction

Over the last decade, typically five to six million tonnes of fly ash has been produced each year from coal-fired power stations in the mainland of the United Kingdom (Figure 1). Between 40% to 70% of the fly ash is utilised in beneficial applications and the remaining material is deposited in landfills. A further 600,000 to 1,000,000 tonnes of furnace bottom ash (FBA) is produced each year. FBA is fully utilised, primarily as a lightweight aggregate for concrete blocks, with little material sent to landfill.

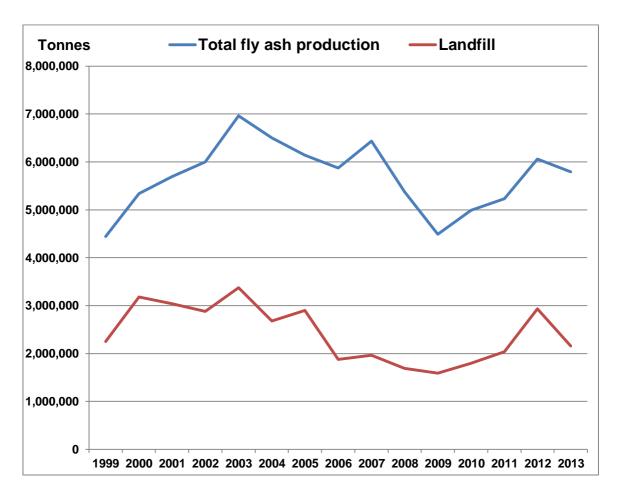


Figure 1 Fly ash production in the UK from 1999 to 2013

2 Uses for fly ash

Uses for fly ash or FBA may be categorised as either bound or unbound. The Environment Agency (EA) in England and Wales consider that bound applications arise if material is "used as an ingredient / component within a product and is fully bound within". Use of fly ash and FBA must comply with the European waste framework directive (WFD). A material considered to be a waste must be recovered and achieve end of waste (EoW) status before it may be used. Achieving EoW status is determined by the regulatory bodies of the member states and should "provide a high level of environmental protection and environmental and economic benefit".

Since October 2010 bound applications for fly ash and FBA have been covered by a Quality Protocol ²(QP), which provides workable EoW criteria. Key aspects of the QP include that the material complies with an approved product standard or agreed specification and has a designated bound application, which for fly ash are:

- Type I addition to concrete (filler or lightweight filler aggregate).
- Type II addition to concrete (cementitious component).
- Cement manufacture e.g. kiln feed.
- Ceramic tiles and brick making.
- Paints, plastics and rubber.
- Lightweight filler for bitumen bound materials.
- Hydraulically bound mixture in pavement construction e.g. road base.

The EA is concerned that unbound applications have greater potential to cause environmental harm, particularly affecting groundwater. Currently, a regulatory position statement ³(RPS 172) exists for unbound applications for a single project up to a limit of 100,000 tonnes, "where the wastes are suitable for use in construction and meet the relevant civil engineering standards for use". There are specific requirements to protect local groundwater and environmentally sensitive locations, such as a Site of Special Scientific Interest (SSSI). RPS 172 will be reviewed during the first half of 2015 once results from an experimental project are available.

With an unbound use for fly ash over 100,000 tonnes a full environmental permit is required. Such a permit often requires significant obligations for site monitoring after completion of the work which may discourage small contractors from using fly ash or FBA in an unbound application, even if justified on technical and environmental grounds.

Fly ash has a wide range of applications within the construction industry (Figure 2). The use of fly ash as a partial replacement of Portland cement in concrete is widespread and considerable volumes are used. Stockpile ash could be a large complementary source but requires a suitable process route to be developed.

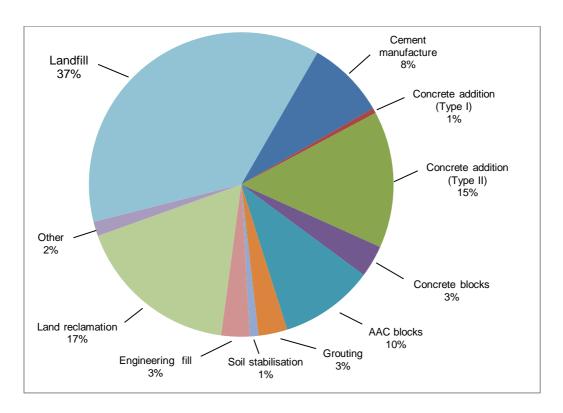


Figure 2 Uses for fly ash in 2013

3 Fly ash use in concrete

3.1 Significance of EN 450

EN 450 ⁴ is the harmonised European Standard for fly ash used as a pozzolan in concrete. Fly ash is a type II addition for concrete in accordance with European Standard EN 206 ⁵ and the complementary United Kingdom standard BS 8500 ⁶. EN 450-1 contains definitions, physical and chemical specifications and the conformity criteria (Table 1). EN 450-2 describes the quality control procedures covering topics such as sampling, autocontrol, auditing and certification. To be considered EN 450 fly ash, material must comply with both parts of the standard. EN 450 fly ash is a recognised constituent used in structural concrete. With a buoyant UK construction sector demand for EN 450 fly ash is increasing.

Property	Limits
Loss on ignition (LOI)	Category A ≤ 5.0%; Category B ≤ 7.0%; Category C ≤ 9.0%
Fineness (45 µm)	Category N ≤ 40.0%; Category S ≤ 12.0%
Free calcium oxide	≤ 1.5%
Reactive calcium oxide	≤ 10.0%
Chloride	≤ 0.10%
Sulfate content	≤ 3.0%
Particle density	± 200 kg/m ³ from stated value
Activity index	≥75.0% (28 days) ≥85.0% (90 days)
Reactive SiO ₂	≥25.0%
$\Sigma SiO_2 + Al_2O_3 + Fe_2O_3$	≥70.0%
Alkalis	≤ 5.0%
Magnesium oxide	≤ 4.0%
Soluble phosphate	≤ 100 mg/kg
Total phosphate	≤ 5.0%
Initial setting time	≤ 200% of cement control
Water requirement	≤ 95.0% of cement control (Category S only)
Soundness	≤10 mm (if free lime exceeds ≤ 1.5%)

Table 1 EN 450-1 specifications

Inclusion of EN 450 fly ash in concrete as a partial replacement for Portland cement gives water reduction, improves cohesiveness of the wet mix and lessens the tendency to bleed. The setting concrete generates less heat from the exothermic reactions, which minimises the temperature gain within the poured mass. Thermal stresses are lower which reduces the incidence of cracking; particularly important if a large volume of concrete is required for a single pour. The specified compressive strength will be achieved in 28 days and will generally increase, due to the pozzolanic reaction between the fine ash particles and lime. The ultimate compressive strength is usually greater than for concrete made without fly ash. Low porosity concrete is produced which reduces migration of chloride ions and other deleterious species through the matrix.

A prime environmental benefit of using fly ash is a reduction in the amount of Portland cement used. Cement consumes a considerable amount of energy during manufacture and the calcination of limestone or chalk liberates substantial quantities of carbon dioxide (CO₂) directly into the atmosphere. Fly ash has low embodied CO₂ and low energy associated with its production. Concrete made with a substantial replacement of Portland cement by fly ash has low embodied energy and a reduced carbon footprint compared with conventional concrete. Less calcareous and siliceous raw material needs to be quarried and fewer fly ash loads sent to landfill, thereby improving resource efficiency.

Portland cement, or CEM I in European terminology, typically has 913 kg CO_2 e / tonne associated with its manufacture ⁷ but the comparable figure for fly ash is only 4 kg CO_2 e / tonne. A typical composite cement produced to European standard EN 197-1 with 30 % fly ash has 655 kg CO_2 e / tonne linked to its manufacture. Concrete produced with the composite cement will therefore have lower embodied carbon than a mix which contains Portland cement as the sole binder. Concrete which contains fly ash is durable and provides a long service life. The technical and environmental benefits of fly ash as a partial cement replacement in concrete are summarised in Table 2.

Benefit		
Technical	Environmental	
Water reduction	Resource efficient	
Greater cohesiveness	Reduced landfill	
Reduced temperature rise	Lower embodied energy	
Lower porosity	Reduced CO ₂	
Lower chloride permeability	Durable concrete and long service life	
High final strength		
Increased resistance to ASR		
Increased durability		
Improved surface finish		

Table 2 Benefits of fly ash use in concrete

3.2 Processing and selection methods

The chemical composition and mineralogy of fly ash is determined by the coal source(s), combustion conditions and cooling rate of material exiting the boiler within the flue gases. If a fly ash meets the chemical specifications of EN 450-1, its performance as a type II addition in concrete is largely determined by the amount of unburnt carbon and fineness. EN 450-1 has two fineness categories and three categories based on loss on ignition (LOI). Processing normally involves reducing the amount of unburnt carbon, as measured by LOI, or increasing the fineness of the output fly ash.

Often most of the unburnt carbon is relatively coarse. For a minority of fly ashes high carbon levels are associated with the fine particles. Size fractionation by sieving or air classifying will indirectly influence the amount of unburnt carbon in the fly ash. Carbon reduction will affect the chemistry and mineralogy of fly ash. This is largely a dilution effect and removal of carbon increases the proportion of the mineral phases such as aluminosilicate glass, generally increasing the Σ SiO₂+Al₂O₃+Fe₂O₃value.

Ten coal-fired power stations operate in the mainland of the United Kingdom. Six of these sites achieve fly ash that meets the specifications of EN 450-1 and conformity criteria of EN 450-2. One of these power stations achieves the required properties by testing, selecting and isolating "run of station" ash of the required quality. Another station in addition to these procedures operates size classification. Three STI separation units operate in the UK and use electrostatic beneficiation to achieve carbon reduction. Of these two STI units can also feed a classifier if further particle size reduction is required. Uniquely at a single power station most of the

carbonaceous material occurs in the coarse fraction of the fly ash. To exploit this property a beneficiation unit was recently installed which uses ultrasonic sieving to achieve carbon reduction and particle size reduction simultaneously.

4 UK coal-fired power stations

A longstanding aim of successive governments has been lower emission of greenhouse gases such as CO₂ from power generation. A 32% reduction in CO₂ emissions from UK energy supply was achieved from 1990 to 2013 ⁸. Less reliance will be placed on fossil fuels such as coal, with emphasis on renewable sources such as wind, tidal and solar; perhaps augmented by new nuclear power stations.

There are challenges for the coal-fired power stations and uncertainty regarding their future. The Industrial Emissions Directive (IED) 9 will place stringent limits on the emissions of nitrogen oxides (NO_X), sulfur dioxide (SO₂) and dust from large combustion plants including coal-fired power stations. Utilities will be required to install new abatement procedures which meet the best available technology (BAT) or otherwise close. Power companies operating coal-fired stations are currently reviewing options. Clearly there are pressures on coal-fired units and it is likely that some closures will occur in the next decade.

Of concern within the construction industry is that perhaps several UK coal-fired stations will soon close, abruptly reducing the supply of EN 450 fly ash. However, coal combustion still accounted for around 32% of power generated in the UK during the period March 2014 until the end of February 2015 ¹⁰. The alternative lower carbon sources of power currently have insufficient capacity to replace the contribution from coal. Installation of any new generating capacity will take several years. In December 2014 the UK Government held a capacity auction to ensure that sufficient generating capacity is available to the national grid for severe weather events in winter, requiring up to 50 GW peak power. Several coal-fired power stations received contracts for 2018 to 2021 under the scheme which means that payments will be made for the units to be on standby during the winter. This scheme provides financial incentives for the power companies to retain coal-fired stations.

5 Fly ash availability

During the summer of 2014 there was reduced availability of EN 450 fly ash. There was concern that this heralded a general decline in the amount of fly ash available to the construction market but the scarcity was actually brought about by three factors. Increased economic activity caused high demand for structural concrete. Additionally, the normal seasonal mismatch of output and demand was operating. Most fly ash is produced in the winter but maximum consumption occurs in the summer. This creates a logistical challenge since EN 450 fly ash is stored and delivered dry and there is a finite volume of silo storage available. However, the most significant factor was the unexpected fall in gas prices. Wholesale gas prices in the spring and summer of 2014 reached historical lows. Several power companies took the opportunity to minimise cost of generation by burning more gas than originally planned. The problem was most acute from June until the end of August. During September combustion units returned from planned maintenance outages, more coal was burnt and the amount of EN 450 fly ash available increased.

6 Potential of stockpile ash

6.1 Conditioned ash

Fly ash is conditioned by mixing with a controlled amount of water (8 to 15% w/w) and discharged into tipper trucks. Conditioned ash is the required form for many geotechnical applications such as engineering fill. Indeed, careful control of the moisture content is critical to achieving the optimum compaction required on site. For large fill contracts a specific stockpile of conditioned ash is often built up over the winter months to ensure uninterrupted delivery during the spring and summer.

Conditioned ash is held in large ash fields throughout the UK. Fly ash may also be slurried with water and pumped to lagoons. Periodically, the lagoons may be drained and the de-watered ash sent to the ash fields or for disposal at a landfill site operated by the power station.

Until the middle of the 1990's most of the UK's electrical power was generated by coal-fired power stations. Consequently large quantities of fly ash were produced but with utilisation rates generally below 50% the surplus material was stored in ash fields or deposited in landfill. UK ash fields may contain up to 50 million tonnes of stockpile ash and this is a large potential source of raw material for use in construction products. If significant amounts of this material could be processed successfully it would augment the supply of "fresh" dry fly ash produced at coal-fired stations and be a complementary source of pozzolan for the manufacture of concrete. The demand for EN 450 fly ash as a type II addition is typically 800,000 to 1,000,000 tonnes per annum. Several decades of EN 450 fly ash could be supplied to the market if a substantial quantity of the stockpile ash was utilised.

The UKQAA has sponsored a research project at the Concrete Technology Unit (CTU) of the University of Dundee to investigate processing methods to allow UK stockpile ash to be used as a pozzolan in concrete. Experimental work started in September 2014 and investigates the reactions and changes that fly ash undergoes when stored moist as stockpile ash. How reversible are the effects and can the original performance of the fly ash be recovered? This knowledge will guide the selection of the most appropriate industrial processing methods to use for recovery. The aim is to develop a process route which is capable of transforming stockpile ash into EN 450 fly ash.

An inventory of the ash fields and landfill sites across the mainland UK is being produced. This will allow an estimate of the total amount of usable fly ash to be determined. Any obvious technical or regulatory constraints will be identified, such as limits on environmental permits at specific landfill sites. How much of the 50 million tonnes can be extracted and processed?

6.2 Recovery of stockpile ash

Recovery is the extraction of stockpile ash, either from lagoons or ash fields, followed by the use of appropriate processing to achieve the properties required for beneficial use in a construction product. Processing stockpile ash occurs for applications such as engineering fill and grouting. This involves removal of lumps and size grading using power screens located at the ash field (Figure 3).



Figure 3 Ash field - United Kingdom (Source E.ON)

Stockpile ash is not used for the production of pozzolan for concrete. There is no commercial process operating in the UK to turn stockpile ash into a dry pozzolan which satisfies the specifications of EN 450-1. Processing methods are outlined in this paper which may be useful in the recovery of stockpile ashes. Several methods may need to be combined in order to achieve a successful process route.

The reactions of fly ash in contact with water are complex and significant chemical and physical changes occur within conditioned ash deposited in ash fields for periods of months to several years. These weathering processes affect the performance of fly ash and limit its use as a pozzolan for concrete. Fly ash is largely composed of spherical aluminosilicate glass particles, but with small quantities of crystalline phases such as quartz, mullite, and magnetite. Adverse changes to aluminosilicate glass particles affects fly ash as a pozzolan. Salts containing sodium, potassium, calcium, magnesium, sulfate, carbonate and hydroxide ions occur, particularly as surface deposits. Much of these salts will be dissolved as water percolates through the stockpile ash. Changes in the bulk elemental composition of stockpile ash are small but there may be some reductions in the amount of sodium, potassium, magnesium and sulfur because of loss of soluble salts.

Weathering of the stockpile ash has two important consequences. Many of the smaller particles become agglomerated together and there is a reduction in the total surface area. Reaction products are deposited on the ash particles and act as a barrier to the pozzolanic reaction, probably by inhibiting the dissolution of aluminosilicate glass. Hydrated calcium sulfates such a gypsum form rapidly in stockpile ash. There is evidence for the formation of calcium carbonate by interaction with carbon dioxide in the atmosphere or from microbial activity. Prolonged weathering may lead to conversion of the aluminosilicate glass into clay-like zeolites ¹¹. Such profound chemical changes are likely to prevent the stockpile ash being recovered successfully.

McCarthy et al ¹² assessed the feasibility of using conditioned ash to produce concrete. Dry fly ashes from UK power stations were conditioned with water and

stored for 18 months in 2 m high stockpiles. Bulk elemental compositions did not change, but two calcium sulfate hydrates, gypsum (CaSO₄.2H₂O) and bassanite (2CaSO₄.H₂O) were significant reaction products. The concrete mixes produced with the conditioned ashes had similar workability to those made with dry fly ash. Generally, the conditioned ashes gave concrete with comparable strengths to controls. However, the fly ash which contained the most lime gave a significant drop in compressive strength after 18 months storage. This showed extensive interparticle bonding in SEM images.

6.3 Processing methods

6.3.1 Blending and sieving

Blending plants for dry fly ash have been built in continental Europe. Some have sieving facilities to control fineness of the product. Fly ashes may be accepted from different coal-fired stations and blended to obtain EN 450 compliant product.

6.3.2 Thermal beneficiation

Stockpile ash was beneficiated using a staged turbulent air reactor (STAR) based in Maryland, USA 13 . Material was screened and fed into the unit without pre-drying. Stockpile ashes with moisture contents up to 30% were processed, at LOI values from 8 to 19%. Combustion was self-sustaining and additional fuel was not required in order to run the unit. To maximise energy recovery from the residual carbon the STAR unit was integrated with the power station. Any combustion based process must take account of the negative environmental impacts of CO_2 , NO_x and SO_2 emissions.

6.3.3 Hydraulic processing

Hydraulic processing of stockpile ash was investigated by Jones et al ¹⁴ who reported a study of stockpile ash obtained from UK coal-fired power stations. Froth floatation was used to reduce the carbon content of slurried fly ash and lamella hydraulic classification produced several size fractions. The processed fly ashes when tested in concrete gave improved consistencies, but no increases in compressive strength were noted compared to the unprocessed stockpile ash.

The Rocktron process combines froth flotation, magnetic separation, the use of hydrocyclones, de-watering and drying to obtain several products from stockpile ash. The major products are a carbon rich fraction, magnetite and a pozzolan comparable to EN 450 fly ash. A full scale production unit is based at the Fiddler's Ferry Power in Cheshire, but is currently mothballed. A pilot plant facility is still operational at the Gale Common site.

6.3.4 Drying units

Drying is an important stage and it is planned that this occurs early in the process route since subsequent processing methods will probably require a dry feedstock. Two drying units for conditioned ash operate in France and one in Germany (Figure 4). Efficient drying must be achieved in order to minimise CO₂ emissions.



Figure 4 Lünen drying unit - Germany (Source SEG/ECOBA)

6.3.5 Electrostatic beneficiation

Electrostatic beneficiation is well established in the UK with three operational STI units. These exploit the difference in electrical charge between the mineral particles which include the aluminosilicate glass and carbonaceous particles to achieve carbon reduction (Figure 5). Inside the separator, the feed ash passes between parallel planar electrodes. Inter-particle contact and friction causes the particles to become electrically charged. The positively charged carbonaceous particles and negatively charged mineral particles are attracted to opposite electrodes and swept away on a moving belt. At one end of the separator the output is low carbon fly ash and at the other end a high carbon material is collected.

Baker et al¹⁵ reported an interesting development regarding electrostatic beneficiation. Two samples of dried and de-agglomerated stockpile ash were processed successfully through an STI separator. Reductions in LOI values from 9.8 to 3.3% and 6.9 to 4.5% were achieved, but surprisingly there was a reversal in the polarity of the charges carried by the particles. The mineral particles were positively charged whilst the carbonaceous particles were negatively charged. The mechanism of the charge reversal is unclear, but apparently efficient carbon reduction may still be achieved.



Figure 5 STI unit - United Kingdom (Source ScotAsh)

6.3.6 Grinding, milling and "mechanical activation"

Grinding dry fly ash in ball mills, vertical mills, vibratory mills etc. has been attempted by several researchers. It is claimed that such processes gives "mechanical activation" and increases the pozzolanic activity of the fly ash. This can be attributed to an increased surface area and perhaps the formation of new reactive surfaces.

The Flubet process ¹⁶ has been developed in Poland for treating fly ash from fluidised bed boilers. "Mechanical activation" is achieved by the formation of micro cracks within granular particles. No reference was found to processing of fly ash from the combustion of pulverised coal. Megapor A is another method claimed to achieve "mechanical activation".

6.4 Process route

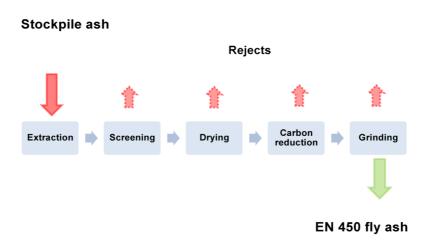


Figure 6 Process route for recovery of stockpile ash

Figure 6 is a schematic of a possible process route to transform stockpile ash into EN 450 fly ash. It envisages extraction and screening in a similar manner as is now required for engineering fill. The third stage is drying to obtain a free-flowing material. Carbon reduction, possibly by a burn-out method or electrostatic separation, is the fourth stage. The final stage is grinding of the fly ash to increase pozzolanic activity by "mechanical activation".

Under certain circumstances the first three stages may only be required to achieve successful recovery of stockpile ash. For example, if a low-carbon conditioned ash has been stored for only a few weeks. Alternatively, it may be more effective to combine the third and fifth stages within a dryer/pulveriser. The process route must be optimised by selection and integration of the most appropriate industrial equipment. Total energy inputs needs careful monitoring to ensure high efficiencies, particularly in the drying and grinding stages and the scheme must be cost effective. It is important to minimise the additional embodied CO₂ associated with the process route otherwise the sustainability benefits of EN 450 fly ash will be reduced.

7 Conclusion

Considerable quantities of fly ash and FBA are produced from UK coal-fired power stations. Both by-products have a wide range of beneficial uses within the construction industry.

There is a strong demand for EN 450 fly ash as a pozzolan for concrete. This by-product will continue to be supplied directly from coal-fired power station sites. There is an opportunity for stockpile ash stored across the UK to be a large source of complementary raw material. The challenge is to develop an effective process route to transform stockpile ash into EN 450 fly ash. A research project is underway at the Concrete Technology Unit, the University of Dundee, sponsored by the UKQAA, to develop suitable techniques.

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Acknowledgements

Dr Hans-Joachim Feuerborn of ECOBA for valuable information regarding European fly ash processing plants.

Prof. Rod Jones, Dr Mike McCarthy and Mr Thomas Hope of the University of Dundee for their support for The Innovative Processing of Stockpile Fly Ash Project.

Colleagues at the UKQAA.