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The Technological and Environmental Implications of Processing Coal Derived Fly Ash in Single Use Deposits

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ABSTRACT

The global environment is very much under the spotlight and the need to reduce greenhouse emissions is having a profound impact on just about every industrial sector. The construction industry is no exception and has been particularly impacted by the energy intensive nature of many of its activities and products.

Coal derived fly ash (CDFA), from the burning of pulverised coal in power station boilers, has long been accepted within the construction sector for the unique properties it imparts to concrete and mortars and as a secondary aggregate in autoclaved aerated block manufacture and grouts. With a goal of zero net carbon by 2050 and the drive to reduce the consumption of virgin materials, the environmental benefits of CDFA are considered alongside the technical benefits. With coal-fired power production expected to cease in the UK by 2024, and for most other European countries by 2030, fresh supplies of CDFA will cease to be available in Europe. As a consequence, future demand will need to be met through imports from countries still operating coal-fired power stations together with the extraction and processing of CDFA from stockpiles.

The UK Quality Ash Association has identified >100 million tonnes of CDFA stockpiles located at currently operating and recently closed coal-fired power stations. The long-term strategic value of these stockpiles for the construction sector has been acknowledged by the UK Government with the need to safeguard the CDFA deposits in the latest edition of The National Planning Policy Framework. Whilst UK stockpiles of CDFA have readily been accepted for use as an aggregate, its potential use as a supplementary cementitious material (SCM) has yet to be realised. In this context, the UKQAA has been working with the Concrete Technology Unit (CTU) at Dundee University and technology providers to investigate the properties of processed deposits of recovered CDFA. Initial findings indicate that processed CDFA can meet EN450 and ASTM C618 specifications. Despite the additional energy required for drying and processing, it is shown in a desk exercise that every tonne of CDFA used as a SCM should still result in a saving of up to 760kg of CO_2 together with some 1.6 tonnes of virgin raw materials (limestone, clays, and shales) when compared to Portland cement (PC) manufacture.

Keywords: Sustainability, Anthropogenic, Circular Economy, Embodied CO_2, Coal Derived Fly Ash, SCM, Beneficiation Technologies, Carbon Removal, De-agglomeration, Properties and performance of CDFA

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1 Introduction

Evolution of coal-fired power production

The UK Government's decision to move away from coal-fired power generation together with the implementation of carbon taxes to reduce the economic viability of coal has resulted in a precipitous drop off in power generated from coal since 2012 (Figure 1). Even power stations that had invested in the latest environmental abatement processes have been prematurely closed leaving only three currently operating stations. All coal-fired power generation is likely to cease by 2024, one year in advance of the original 2025 deadline (UK Government, 2021).

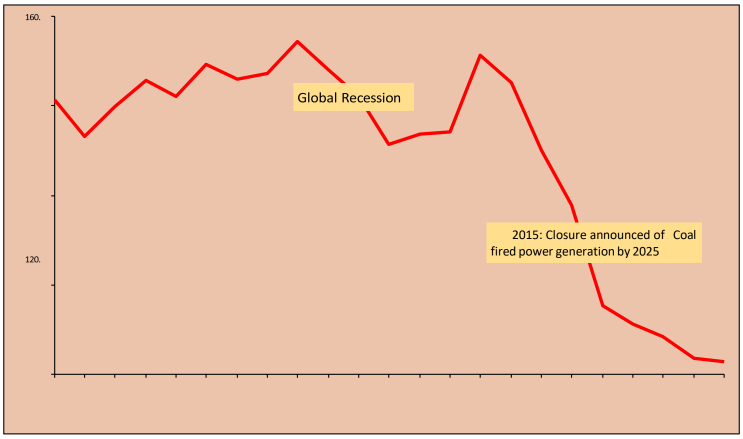

Figure 1. UK Coal-fired Power Generation 1998 to 2020

Implications for Coal Derived Fly Ash supply

Figure 2 demonstrates how the move away from coal-fired power production has impacted on coal derived fly ash (CDFA) availability. If seasonality is taken into account, the impact is even more pronounced with coal-fired generation only tending to be required during the winter period. With markets not able to rely on CDFA arising from UK generation, this has placed an emphasis on identifying import sources, as well as exploring options for the extraction and processing of CDFA that has been deposited in stockpiles over the past 30 to 40 years (DBEIS 2017). The other option in DBEIS (2017) is through performance specification for CDFA in Portland cements and concretes.

Figure 2. Generation of CDFA

Safeguarding Stockpiles of Coal Derived Fly Ash

The UKQAA estimates that stockpiles of CDFA at currently operating or recently closed coal- fired power stations are in excess of 100 million tonnes (Figure 3) with a reasonable spread across the country. It should be noted, however, that the deposits can differ in quality and ease of access.

Figure 3. UK Power Stations

The key is how to safeguard these stockpiles for future generations and this needs to start with accurate definitions that can be included within local and national planning policy documentation. The definition: "coal derived fly ash (CDFA) in single use deposits" allows for fly ash from coal- fired power generation to be differentiated from incinerator and bio-mass fly ashes which can have very different physical and chemical properties. The term "single use deposits within defined boundaries" helps further to avoid confusion with general purpose landfill sites which may include other wastes or contaminants. The UK is fortunate that CDFA is not contaminated with 'bottom ashes' from the boilers as there are well established markets for this material in blocks.

CDFA recovered from stockpiles from coal-fired power production is referred to as a 'secondary aggregate' in planning terminology. In reality, one could argue that it can be defined as a 'secondary aggregate' or 'secondary mineral' depending on the eventual application (see Section 2 below).

The Ministry for Housing, Communities and Local Government (MHC&LG) has recognised the need to safeguard the existing 'single use deposits' of CDFA for the construction sector and CDFA is referenced in the latest revision of The National Planning Policy Framework (https://www.gov.uk/government/publications/national-planning-policy-framework--2022). Whilst this does not guarantee the future availability of every stockpile of CDFA, it ensures that planning authorities must take such deposits into account when looking at alternative options for the site.

2 UK market for coal derived fly ash

2.1. Evolution of supply-demand

The evolution of the supply-demand for CDFA together with a medium-term outlook based on its potential use as a secondary aggregate and SCM is shown in Figure 4.

Figure 4. Supply – Demand Balance for CDFA

In 2012, of the six million tonnes of fly ash generated in the UK, some 50% was landfilled in single use deposits resulting in the utilisation of around 3 million tonnes. A significant proportion of this tonnage was, however, used in road sub-base and low value fill applications.

As CDFA availability from power generation declined, in order to meet demand, supplies of 'fresh' CDFA were augmented by imports and extraction from power station stockpiles. The changes in supply – demand balance have precipitated an inevitable increase in prices and a subsequent move away from low value fill applications to higher value-added markets. Moreover, the level of consumption depicted in Figure 4 is likely to be much lower than the potential demand for CDFA which is currently constrained by lack of supply.

With coal-fired power generation coming under increasing environmental pressure, there is likely to be little appetite within the global capital communities to support any future investment in pulverised coal-fired power generation. As a consequence, securing long-term sources of good quality CDFA will become increasingly strategic and competitive. This will include technologies to assist in its extraction from legacy stockpiles although, it should be noted, that not all deposits are technically or commercially viable for recovery.

2.2. Cementitious markets

The use and benefits of CDFA for applications in cementitious markets has been well documented. Whilst durability, workability, pumpability, long-term strength gain and low heat generation in mass pours continue to be key factors driving demand, (Concrete Society, 2011), of more recent interest is for use in low embodied carbon applications.

The increasing cost of CO_2 (Figure 5 below) coupled with pressure for more sustainable solutions and durable structures with >100 years service life offers significant growth opportunities for CDFA. Indeed, there are virtually no concrete or mortar applications that would preclude the use of CDFA in cementitious applications. Subject to the Portland cement chemistry, quality of CDFA and the application, we may well start to see higher Portland cement replacement levels, standards permitting.

Figure 5. Value of EU Carbon Permits (€)

Finally, it should also be noted that work is currently being undertaken on alkali activation of CDFA (geopolymer cement) as a total Portland cement replacement. Replicating the traditional benefits of ready mixed concrete in terms of handling, ease of placement and commercial viability will, however, present a number of challenges and it is likely that the initial advances will be made with precast concrete products such as bricks, blocks and beams.

2.3. Aggregates Markets

Cementitious Grouts

The inherent spherical shape of the fly ash particles and low density means CDFA is ideal for grouting and ground stabilisation applications where 'flow' is an important characteristic. This is particularly the case when stabilising old mine shafts to avoid subsidence. The grouts may contain between 50% and 90% CDFA with the balance being Portland cement. Annual requirements are very much project and infrastructure related though UK demand in any particular year may range from around 250kt to over a million tonnes.

Autoclaved Aerated Concrete Blocks

Since their introduction in the late 1950s, aircrete products have won rapid acceptance throughout the building industry as a result of their ease of handling, insulating properties and sustainability credentials. (https://www.hhcelcon.co.uk) An autoclaved aerated concrete block plant can require over 200,000 tonnes of CDFA per annum. Aircrete is produced by mixing CDFA with water to create a slurry. This is then pumped into a mixer where binders (Portland cement and lime) are added, along with a small quantity of aluminium powder which reacts with the water to create the lightweight 'aerated' honeycomb bubble matrix. After being allowed to rest enabling the build-up of initial strength, the resulting 'green' material is cut to size and placed into an autoclave to complete the curing process.

Raw material feed for clinker production

CDFA can be used as a shale or clay replacement in the production of Portland cement. Not only does CDFA replace virgin raw materials (including bauxite for the alumina content), it can also help reduce kiln emissions such as sulphur dioxide as sulphur is often present in clays and shales.

Stabilisation

CDFA has historically been used as a constituent to stabilise soils, sludges and wastes to reduce the potential for leachates from landfill sites. When mixed with Portland cement or lime, CDFA can also be used for soil stabilisation with enhanced mechanical properties including the reduction of ‘sulphate heave’ (Britpave 2019). Stabilised dredgings from canals have been recycled by into tow paths.

3 Properties of single use deposits of CDFA

CDFA from pulverised coal-fired power stations is universally accepted for the unique properties it imparts to concrete. From a cementitious perspective, key requirements include:

- The CDFA conforms to European and national standards.
- The chemistry of the CDFA offers cementitious (pozzolanic) properties by reacting with the lime produced from the hydration of Portland cement in concrete or mortars.
- Carbon in the CDFA from the incomplete combustion of coal is known to have a detrimental impact on the properties of mortars and concrete and both European and American standards have limits for carbon content when CDFA is used in cementitious applications.
  - Carbon offers no cementitious properties
  - Carbon has a detrimental impact on air entrainment and chemicals added to improve the performance of concrete
  - The science and activity of the carbon and its effects on concrete additives can be very complex
- The fineness of the ash particles is sufficient to optimise the chemical reactions that are required to create cementitious properties
  - Increase in fineness is normally associated with increased rates of chemical reactions, ceteris paribus.
  - The combination of the CDFA chemistry and fineness has an impact on mortar strength measured at 28 and 90 days. (Referred to as the ‘activity index’).
  - Fineness and particle size distribution also make an important contribution to packing density and subsequent impact on durability of concrete.
- There is a high proportion of glassy spherical particles which allow for flow in concrete and mortars. Often referred to as the ‘ball bearing’ effect.
- The CDFA should be of consistent quality to minimise variability in concrete and mortars.

The UK is fortunate that CDFA stockpiles are defined as ‘single use deposits’. This means that the CDFA should not be contaminated with furnace bottom ash (FBA) or any other types of power station waste such as gypsum, refractory bricks, and flue dusts. A further feature is that UK pulverised coal-fired power stations were designed to burn hard coals which tend to be similar in terms of chemistries (siliceous, low lime) and offer good cementitious properties.

When CDFA is stored in open stockpiles or sluiced to lagoons, water is added to allow for transport and to reduce dust emissions. The addition of water to CDFA can result in both physical and chemical changes to the ash particles. These include the development of inter- particle cohesive forces, sulphate, and carbonate product formation and pozzolanic (cementitious) reactions in the presence of free lime (McCarthy et al 2017). The inter-particle cohesive forces together with the pozzolanic reactions from the presence of free lime result in the ash particles sticking together (known as agglomeration) and resulting in a subsequent reduction in fineness. Such effects can influence CDFA reactivity although research suggests that the material still exhibits pozzolanic properties, with potential for enhancement through processing.

Indicative property ranges for single use deposits of CDFA in the UK, from work undertaken by the University of Dundee in collaboration with the UKQAA, are given in Table 1. While these indicate variability, it should be recognised that properties such as moisture content, fineness (45 µm sieve retention) and loss of ignition (carbon content of the ash) can be modified through drying and technologies that increase fineness (de-agglomeration) and remove the carbon. Physical and chemical properties range of the fly ashes are typical of those produced for the coal type used, with only minor changes tending to occur following prolonged wet storage.

Table 1. Selective properties of fly ash from various stockpiles (% unless noted otherwise)

Property	Range	Mean	Component	Range	Mean
Moisture Content	6.0 – 21.1	15.8	SiO_2	41.2 – 51.2	46.5
Fineness, 45µm sieve ret.	41.1 – 63.2	50.2	Al_2O_3	19.5 – 25.2	22.9
Median Particle size, µm	28 – 44	32.4	Fe_2O_3	5.8 – 9.4	7.9
Loss-on-ignition	3.5 – 15.9	8.5	CaO	2.1 – 4.4	3.0
Water requirement	102 – 109	106.8	Na_2O_{eq}	2.1 – 2.8	2.4
28 days Activity Index	71 – 83	74.6	SO_3	0.8 – 2.3	1.6
90 days Activity Index	80 – 90	84.1	Glass/others	66.0 – 79.2	73.7

The agglomeration as a result of the surface charge/cohesive forces and the nature of reactions occurring on the surfaces of the ash particles after prolonged wet storage means deagglomeration of CDFA to create greater surface area would be necessary. Figure 6 (a) shows a sample of agglomerated stockpiled ash and Figure 1(b) the same sample after lab scale grinding.

It is believed that grinding initially breaks down the agglomerated CDFA particles. At more intensive levels of grinding, the fly ash spheres may also begin to fracture and lose the “ball bearing’ effect. It appears there is an optimum level of grinding with regard to releasing the spheres and ensuring the beneficial influence of fly ash on the fresh properties of mortar or concrete. It has been found that strength continues to improve with extended grinding and increased fineness (McCarthy et al, 2022) though it should be noted that grinding will also increase the fineness of any carbon present in the fly ash which will have a detrimental impact on the effectiveness of chemical admixtures used in concrete and mortars.

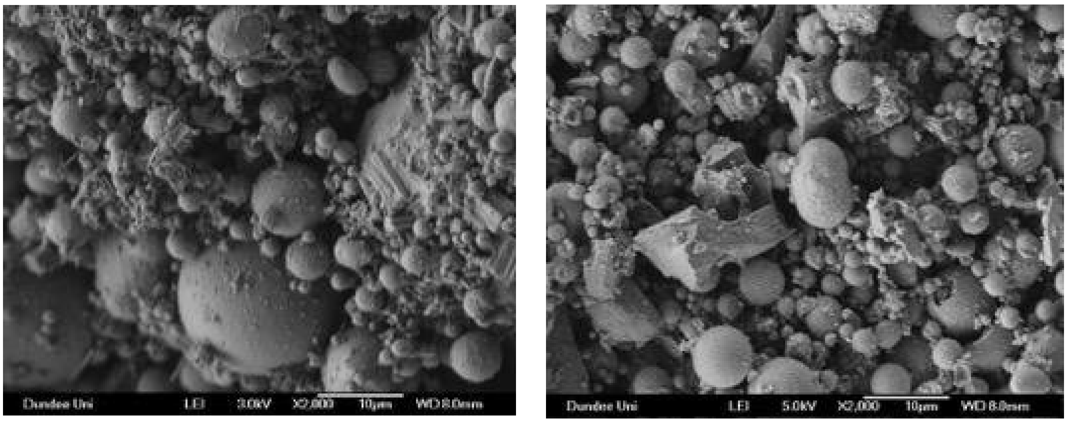

Figure 6. (Left) stockpile fly ash and (Right) following grinding in a ball mill for 20 min. (Courtesy of Dr Thomas Hope, University of Dundee)

An example of activity index and particle size data for two stockpile fly ashes before and after laboratory scale drying and grinding is shown in Table 2. This demonstrates that initially, the fly ashes exhibited values close to the Standard (EN 450) requirements at 28 and 90 days, (75% and 85% respectively). Following grinding for 20 minutes, values above the Standard limits were obtained. These results could be improved further by screening off >600 µm material and extending the grinding time to 120 minutes to further increase the fineness.

Table 2. Activity index and particle size parameters following laboratory processing.

Material	Activity Index, %		Particle Size Parameters	
	28 days	90 days	Median size (d_{50}), µm	Sub 10 µm content, %
As received				
SFA1	74	83	31	21
SFA2	71	86	44	20
Drying/20 minutes grinding - large batch				
SFA1	84	101	14	35
SFA2	78	97	15	35
Drying/< 600 µm/120 minutes grinding - small batch				
SFA1	91	106	5	71
SFA2	91	121	5	74

The data in Table 2 indicates that the quantity of sub 10 µm particles in fly ash (and median particle size) influence behaviour. Wider testing suggests that quantities of around 30% or more of sub 10 µm particles are necessary to pass the activity index test (McCarthy et al, 2022).

It should also be noted that the source of Portland cement used in the activity testing may have an impact on the results due to different Portland cements having subtle, though important differences, in their chemistries. Portland cement chemistry and fineness also have a significant influence on the performance of CDFA in concretes and mortars. In terms of concrete testing, the impact of CDFA fineness becomes more noticeable once the sub 10 µm particle content exceeds about 50% (McCarthy et al, 2018).

As outlined in Section 4 below, there are a number of processing technologies available for carbon removal and the drying and de-agglomeration of stockpiled CDFA. In order to ensure there was sufficient volumes of CDFA for the larger scale testing at Dundee, use was made of the Atritor (combined drying and de-agglomeration) and STET (carbon removal) pilot plants to process the stockpiled ash samples to ensure the CDFA 'conformed' to a minimum of the EN450 B/N standard in terms of fineness and carbon content of the ash.

- At pilot scale level, the CDFA could be dried and deagglomerated to < 0.5% moisture and with a fineness range meeting both EN450 category S and N requirements depending on pilot plant set up.
- The STET pilot plant was able to reduce the carbon levels to ~3.5% with throughput and yield subject to the inherent characteristics of the fly ash deposit.

The key for any commercial operation will be to optimise the drying, deagglomeration and carbon removal to ensure specific physical and chemistry requirements are met at minimal cost.

4 Processing of single use deposits of CDFA

Figure 7 demonstrates the various processing routes that can be used for single use deposits of CDFA subject to the intended application. As a general rule of thumb, secondary aggregate and minerals applications tend to require less processing than for use as a SCM though this is not always the case.

Figure 7. Process Flow for Secondary Aggregates and Cementitious Applications

This section reviews industrial processes, at different stages of development and commercial acceptance, for drying, de-agglomeration and carbon removal.

4.1. Drying Technologies

When it comes to the processing of landfill or lagoon ash, drying and de-agglomeration account for the most energy consumption and this places a premium on identifying efficient solutions. Key considerations include:

- How to minimise the moisture content of the stockpiled ash prior to introducing into the dryer?
    - Windrows to allow for drainage and some air drying.
    - Covered storage.
- Ensuring an even dispersion of fine ash particles into the dryer in order to maximise the surface area for moisture removal.
    - Availability of waste heat sources

Atritor (http://www.atritor.com)

The Atritor process allows for flash drying, de- agglomeration and classification to all take place simultaneously in an air swept mill (Fig. 8).

Figure 8. Air swept mill

Work undertaken by Atritor in the UK demonstrates that it is possible to dry stockpiled CDFA and to de-agglomerate to an EN450 S or EN450 N fineness specification. Table 3 shows the results from the testing of a number of stockpiled ash samples. The resultant products were then sent to STET for carbon removal prior to lab trials at Dundee University.

Table 3. Pilot Scale testing of stockpiled ashes (bag filter collection not measured; likely to be fines <10μ)

	Moisture		**Fineness**	
	Feed (%)	**Output (%)**	**<45μ (%)**	**<10μ (%)**
UK Sources				
Sample A	22	0.5	79	30
Sample B	25	0.4	75	30
Sample D	15	0.5	65	25
Non-UK	*(LOI 10.5% and mix of bottom ash)*			
Sample C	22	0.5	65	20

Flash Dryers

The specification requirement for flash dryers is very much dependent on the material to be dried though they all work on the principle of dispersing a wet granular material into a stream of heated air. The aim is to 'strip' the moisture off the surface of the ash particles and the gas temperature and flow must be sufficient to retain the water to avoid it from re- condensing back onto the ash. Flash dryers are often used in conjunction with hammer mills or flails to break up the ash particles and the dried product can then be captured in cyclones or bag filters.

Coomtech (https://coomtech.com)

Coomtech is a UK-based company that has developed a drying process which is claimed to have reduced energy and CO_2 emissions by up to 75% when compared with traditional thermal drying systems. The innovation, known as Surface Moisture Remove (SMR), uses turbulent air to shear moisture off the surface of particles. The air is warmed to 85°C to ensure the moisture is removed without resettling.

The company has built a pilot plant in Adlington, UK and the technology has been approved by the Solar Impulse Institute. An industrial scale plant is currently being built in Australia to dry coal for hydrogen production.

4.2. Grinding Technologies

Section 3 has shown that, with landfilled fly ash, it is important to de-agglomerate the ash particles to improve ash fineness and the subsequent reactivity.
The Atritor mill offers drying and de-agglomeration simultaneously. Other drying processes, however, might require additional processes to provide a fine dispersion of ash particles into the dryer, as well as ensure that the sub 10 micron fraction is sufficient to meet the activity index requirements.
In this respect, commercially available technologies include:

- Hammer mills and flails
- Vertical mills
- Ball mills
- Booster mills for ultra-fine grinding

The key will be the optimisation in achieving the desired fineness at an economic cost of energy and wear on the mills.

4.3. Carbon Removal Technologies

Section 3 mentioned the importance of reducing the level of carbon in CDFA to limit its deleterious impact on admixtures which are used to improve the performance of concrete and mortars. These include air entrainers and super-plasticisers.

Carbon removal technologies rely of segregating the carbon from the mineral component of the CDFA. This can either be achieved by flotation, mechanical separation or electro-static means. As the carbon is removed from the ash, consideration needs to be given to process yields and whether there are markets for the separated carbon fraction which can range from 25% to 50% carbon.

STET (previously known as Separation Technologies Inc) (https://steqtech.com/about-us/)

The STET process is based on the tribo-electric charging of fine particles with different chemical properties. In the case of fresh CDFA, the carbon normally takes on the positive charge relative to the mineral component thus allowing the carbon to be separated from the mineral content by passing the CDFA between two charged plates. A fast moving belt is used to aid separation and to ‘sweep’ the separated particles to their respective hoppers. The ‘captured’ carbon can be returned to landfill or has the potential to be used in applications such as waste stabilisation, asphalt or a ‘scrubber’ in biomass plants.

The first STET Separator was installed in Massachusetts, USA in 1995 and over the past 27 years, more than 18 million tonnes of fly ash have been processed in the USA, Europe and the Far East. With the focus now on processing landfill CDFA, STET has built a demonstration plant at the Brunner Island power station in Pennsylvania USA to process up to 6t/hr. Several industrial scale plants are at an advanced stage of feasibility studies and negotiations.

SonoAsh (https://www.sonoash.com)

SonoAsh is a wet beneficiation process for carbon removal from CDFA. The key component of the SonoAsh technology is the “low frequency sonic reactor” which is stated to simultaneously separate out the carbon and to de-agglomerate the ash particles. The CDFA is introduced into the reactor in the form of a wet slurry and by implication, the resulting fractions will need to be separated by flotation or hydro-classifiers and then dewatered and dried. The technology has yet to be introduced on a commercial scale.

Sieving/Classification

Sieving technologies to remove carbon from the CDFA have been installed at a number of power stations over the years though it is not known if any are still operating commercially. The key requirement is for carbon to be present in the coarse fraction. Experience suggests that this cannot be guaranteed so sieving has had limited success. Maintenance of the screens and the risk of blinding are also key considerations.
Classifiers can be mechanical, or air swept though, like sieving, they rely on the carbon residing in the coarse fraction. If there is a high percentage of fine particles, both classifiers and sieving can actually increase the LOI in the final product.

Rocktron

The Rocktron process relies on being able to separate the carbon from the mineral ash through flotation. A commercial scale facility was built at Fidlers Ferry power station in the UK though, for a number of commercial and technical reasons, it was mothballed.

4.4. Carbon Burn Out Technologies

Carbon burn out processes rely on heating the CDFA to temperatures in excess of 700°C in the presence of oxygen to convert the carbon to carbon dioxide. The key is to control the temperature of the ash to avoid fusion of the mineral component. Carbon burn out technologies need to factor in the emissions to air and the requirement for environmental abatement equipment. A further consideration is that the conversion of carbon to carbon dioxide releases almost 4 tonnes of CO_2 gas for every tonne of carbon that is removed. Unless carbon capture technology is included in the stack, the resultant CDFA will carry a higher carbon dioxide weighting.

SEFA (STAR® Technology) (https://www.sefagroup.com)

SEFA operates three STAR beneficiation facilities of which it owns two. According to SEFA, there are three further development opportunities for their technology. The current units are under construction in North Carolina where there is a legal obligation on the Utilities to remove their stockpiled CDFA from existing landfill deposits.

The essence of the SEFA – STAR process is a type of turbulent fluidised bed reactor vessel which is pneumatically fed with pre-dried fly ash. Initially the CDFA would need to be heated by gas though with >6% LOI ash feed, the process can become auto-thermal. At higher levels of LOI, controls might need to be put in place to manage heat generation within the reactor vessel.

Boral (CBO Technology) (https://flyash.com/products-and-technologies/carbon- burn-out/)

The Boral – CBO fluidised bed process was first introduced at Wateree power station in South Carolina in 1999. Historically, four CBO facilities were constructed and have reportedly processed in excess of 7 million tonnes of ash although none are believed to still be in operation.

Whereas the SEFA-STAR process uses a turbulent reactor vessel operating at high temperature, the Boral – CBO process uses “dense phase” or “bubbling bed” technology. The aim is to maintain the bed at around 700°C to burn out the carbon without overheating and causing fusion of the ash particles. Boral is currently looking at how their technology can be adapted to receive landfill ash.

Charah (MP618™ Technology) (https://charah.com)

The Charah ash beneficiation process is an adaptation of SCB’s patented mercury removal technology. Charah has a small pilot plant operating in Louisiana, USA and the company has stated that full scale plant is being planned. The process comprises a steel tubular rotary shell which can be heated on the outside by a series of burners located around the circumference and along the length. The tube is positioned at a slight angle which allows the fly ash to flow through it - aided by gravity and a series of paddles. When the fly ash reaches some 700°C to 750°C, the carbon is burned off. Process control is a critical factor to avoid overheating of the shell.

4.5. Summary

Over the past 20 years, ash beneficiation requirements have led to many innovative ideas though relatively few have made it through to commercial reality. However, should there be a requirement to beneficiate stockpiled CDFA, there is a choice of commercially proven technologies available.

5 Environmental benefits of using fly ash

5.1. CDFA and the Circular Economy

The main premise of the Circular Economy is to move away from linear product life cycles to ensure that the 'end of life' for products or infrastructure allows for recycling. The aim is to reduce consumption of virgin raw materials and, as a consequence, the volume of waste sent to landfill. Anthropogenic - the impact of human activity on the environment – is a word that is becoming more closely associated with the circular economy and focus on industrial processes which minimise their impact on the environment.
There are three ways in which CDFA can enter into the circular economy model.

- The first is to ensure that any CDFA from the limited coal-fired power production is collected and used in cementitious or secondary aggregates markets rather than directed to landfill.
- The second is to maximise the utilisation of 'legacy' stockpiles of CDFA. These stockpiles can often be used 'unprocessed' as a secondary aggregate or it can be processed to meet the physical and chemical needs for cementitious markets.
- The third, and perhaps the purest link to the circular economy, is the recycling of concrete and manufactured building materials which contain CDFA.

5.2. Sustainability benefits

From an environmental perspective, every tonne of 'fresh' CDFA captured in the power station precipitators and used in Portland cementitious applications saves some:

- 1.6 tonnes of virgin materials such as limestone and shale
- 860 kg/tonne CO_2

For stockpiled CDFA, one needs to take account of the processing energy prior to arriving at a net CO_2 benefit. Assuming a moisture content of 20% to be reduced to <0.5% and a carbon level of 12% reduced to <5%, there is a net benefit of:

- ~760 kg/tonne for 'carbon removal' process technology
  - Processing accounts for around 100 kg CO_2/t
  - ~410kg/tonne for 'carbon burn-out' technology
  - Processing by burn out technologies release almost 4 tonnes of CO_2 per tonne of carbon removed.
  - The figure is very sensitive to the level of carbon in the feed ash. Normally the resultant product has <2.5% carbon.

When the CDFA is used as a secondary aggregate and substitutes for sand in grouts and concrete products, every tonne of CDFA saves:

- >1 tonne of virgin sand based on the relative density of the two materials.
- There may be also some CO_2 benefits subject to the source of sand. Marine dredging will have a higher CO_2 footprint as well as a requirement for fresh water to remove chlorides.

5.3 CO_2 Allocation between Primary Products and By-Products

Understanding how a product or industrial process impacts on the environment should be welcomed if it enables informed decisions to be made by the various stakeholders. Informed decisions, however, require confidence in the robustness of the assumptions and calculations. These can be extremely complex and often creates many more questions than answers.

"Greenwashing" is now entering the lexicon of everyday language as a result of questionable accounting, reallocation of energy inputs or debatable carbon offsets. Once public confidence is lost in the integrity of the process, it will be difficult to regain. It is also important to ensure that we do not put more resource into the measurement and the accounting of CO_2 rather than the technology and science to reduce the absolute level.

A recent development in the steel industry has been an attempt to justify the reallocation of some of the CO_2 generated by the ironmaking to the blast furnace slag. There are no benefits to the environment with the only aim being to reduce the carbon footprint and taxation on the pig iron and, by implication, the steel. Countless hours and resource have then been spent on ludicrous discussions on whether the CO_2 allocation been the pig iron and slag should be by the relative mass balances or by economic value! What happens if, as a result of carbon tax, blast furnace slag becomes uneconomic? Would any resulting stockpiles be seen as carbon storage from a tax and emissions perspective?

The arguments being made by the steel industry are migrating to other sectors such as coal- fired power generation and ferro-silicon production. It is spawning a new industry of consultants and auditors who would be better focussed on the science and technology.

The premise of this paper is that any CO2 allocation to by-products should only be related to the incremental energy used to process and handle them.

- In the case of CDFA stockpiles, the CO_2 calculation should start with the extraction of the ash and then add the energy associated with any further processing.
- In the case of 'fresh ash' the CO_2 calculation should start at the conditioning silos after the precips as conveying the ash from the precips to the conditioning silos still forms part of the power generation process.

6 Desktop study of the CO_2 implications of processing 250,000 tonnes/yr from stockpiled single use deposits of CDFA

The CO_2 footprint of stockpiled CDFA is dependent on a number of factors:

- Location of the deposit and ease of access
- The moisture level during different seasons of the year
- The level of processing required
    - Does the CDFA require de-agglomeration or grinding to improve fineness?
    - Does the CDFA require to be dried and, if so, are there sources of waste heat?
    - Is there a requirement for carbon removal?
- Availability of 'green energy'
    - Electricity generation
    - Drying gas such as hydrogen fuelled burners

The following 'desk top' calculation is based on discussions that have taken place in the UK with contractors who manage stockpiled CDFA and technology providers – Atritor and STET – who were involved in pilot scale tests to process stockpiled CDFA to supply material for testing by Dundee University.

The process flow used for the basis of the calculations is illustrated on Figure 9, below:

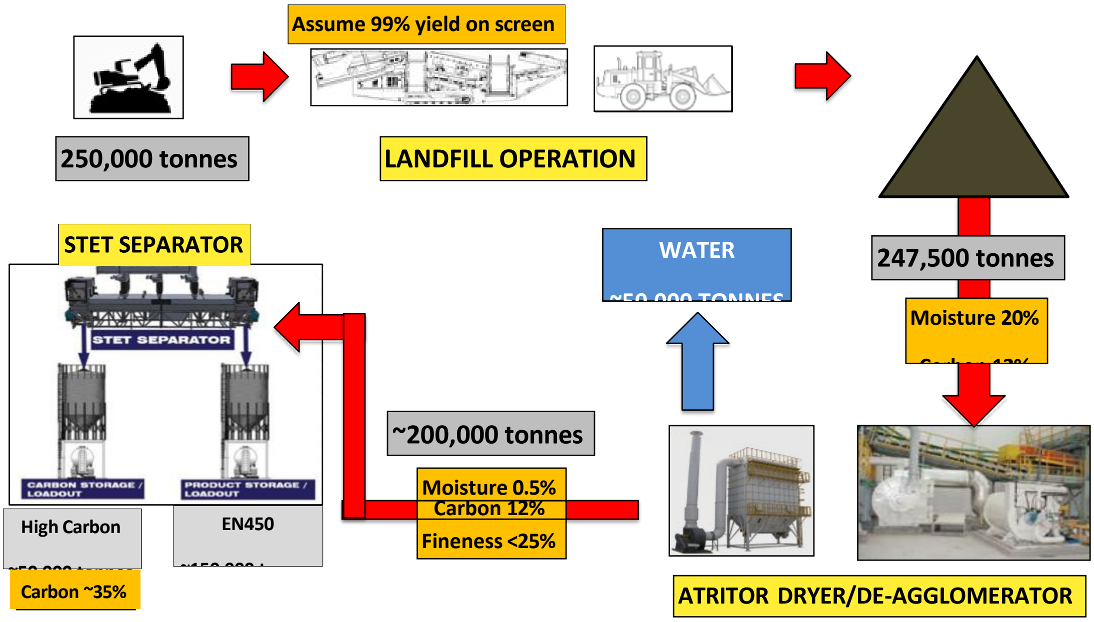

Figure 9. CO_2 Implications of Processing 250,000 tonnes of stockpiled CDFA

6.1. Landfill Stockpile Extraction

The landfill operation is assumed to require bulldozers, shovels, screens, dumper trucks and water bowsers. With single use deposits of CDFA, the yields are expected to be very high once organic material is scraped off the top layer which is why a recovery figure of 99% is used.

It is recognised that every stockpile of CDFA will have its own idiosyncrasies and the fuel consumption will be site and plant dependent. Whilst these factors will impact on CO_2 emissions, of greater interest will be how plant operators foresee the future energy sources for powering their plant. Fuel cells? Bio-fuels? Hydrogen gas? In this context, JCB has recently announced that it has been investigating the potential to use hydrogen gas to power its plant and machinery. https://www.jcb.com/en-gb/news/2020/07/jcb-leads-the-way-with-first-hydrogen-fuelled-excavator

If hydrogen is adopted as the main source of power, then the CO_2/tonne of CDFA extracted would fall to close to zero. This assumes that there is no amortisation of embodied CO_2 in the fabrication of the actual mobile plant and machinery.

Key assumptions for landfill extraction are provided in Appendix A1. The figures included in the Appendix have been based on feedback from a contractor specialising in operating with CDFA stockpiles. The assumptions have been included to enable understanding and to look at sensitivities based on other stockpile experiences.

6.2. Drying and De-agglomeration (Atritor)

Drying and de-agglomeration is by far the most intensive use of energy: both in the form of electrical energy for processing and de-agglomeration and the gas used for drying. This might not be required if the CDFA is used as a secondary aggregate though for use as an SCM, the stockpiled CDFA needs to be dried, de-agglomerated and possibly subjected to carbon removal.

For the purposes of the case study, Figure 7 assumes a moisture content of 20% and carbon level of 12%. Anything that can be done to bring the moisture level down by pre-drying in'windrows' or allowing to drain under covered storage will have significant benefits on thedrying costs and resultant CO_2 emissions.

The ability to utilise waste heat from other processes or to replace natural gas with hydrogen will also have a significant impact on the CO_2 allocated to the drying. In the case of the electrical power, rather than assume 100% renewable energy, the model has assumed the current UK emission factor: 0.21233 kg CO_2/kWh (www.electricityinfo.org). This value will fall as the UK continues its move towards 100% green energy.

Key assumptions and outputs provided in Appendix A2 were based on the Atritor pilot plant testing with assumptions made for the power requirements for the balance of plant to feed the Atritor mill and to convey the resulting product.

6.3. Carbon Removal (STET)

The desk top study has been based on the feedback from pilot scale tests using the STET tribo-electric carbon removal technology. STET carbon removal technology has a long history of use for CDFA and there is a very good understanding of the power requirements for running a separator including feed, filters and conveying systems (balance of plant).

Whilst the STET tribo-electric carbon removal process uses relatively little energy in its own right, the 'yield' of final product does have a significant impact if the CO_2 generated over the whole recovery process is fully allocated to the final product. The yield is very much a function of the carbon content of the feed ash and the target level for the final product. The higher the level of feed carbon and the lower the desired level of carbon in the final product, the lower the yield, ceteris paribus.

In the case study, the feed level of carbon is assumed to be 12% and the final product target is <5%. Pilot plant testing indicated a yield of 75% is achievable based on the dried samples received by STET from Atritor. It should be noted that experience has shown that stockpiled ashes do not necessarily all exhibit the same behaviour, hence the requirement for testing.

Key assumptions for carbon removal are provided in Appendix A3. The STET separator uses relatively little power and the key variables are the estimated yields and what is required for the 'balance of plant' to feed the separator and then to transport the resulting products to silos. Figures used for the balance of plant are based on STET's extensive experience of designing, building and operating carbon removal plants.

With carbon burn out processes, yield is much less of a factor as there is no by-product. The yield is basically a function of the level of carbon and moisture removed. However, every tonne of carbon removed through burn out technology results in some 4 tonnes of CO_2 gas not including any CO_2 related to the electrical power to run the processing plant (Appendix A4).

6.4. Results from desktop studies

Table 4 (below) summarises the estimates for the CO_2 generated during the various stages identified in Figure 6.1 and then allocated to the final product.

Table 4. Estimates for the CO_2 generated during different stages.

Process	Ash (tonnes)	Yield (%)	CO2 (tonnes)	Split	Product (kg CO_2/t)
Landfill Extraction	250000	99	1620	11.23%	10.8
Drying	247500	80	12231	84.81%	81.9
Processing	199238	75	571	3.96%	3.8
Final Product	149428		14422	100.00%	96.5

- Feed CDFA: 250,000 tonnes
- Product CDFA (EN450 equivalent): 149,428 tonnes
- Overall yield 60%
- Total CO_2 produced: 14,422 tonnes
- Total CO_2/tonne product: 96.5 kg/tonne

It is recognised that any desk top study has its limitations and that every CDFA stockpile and processing plant design will be unique to the location and local environmental factors. However, with drying accounting for >80% of the CO_2 generation, it is clear that this needs to be the focus of attention. In this respect, key elements include:

- Use of windrows and covered storage to reduce the moisture content of the feed ash
- Maximising use of waste heat from other processes
- Investigating alternative drying technologies
- Replacing hydrocarbon-based gases with hydrogen for drying

7 CONCLUSIONS

The move away from pulverised coal-fired power stations has resulted in shortages of CDFA for use in the construction industry. The UKQAA has identified >100 million tonnes of stockpiled of CDFA in single use deposits in currently operating or recently closed power stations. The strategic importance of these deposits has been recognised by the UK Government and the need to safeguard these stockpiles is now enshrined in national planning policy documents.

Stockpiled CDFA can be used as a secondary aggregate for use in autoclaved aerated block manufacture and grouts for ground stabilisation with minimal processing. For use as a SCM, further processing is required to dry and de-agglomerate the CDFA. Carbon may also need to be removed to comply with EN450 standards.

Work undertaken with the CTU at Dundee University has shown that whilst stockpiled CDFA does not have the same reactivity as fresh CDFA, providing fresh surfaces are created during de-agglomeration and that >30% of the particles are less than 10 microns, the processed ash can meet EN450, with potential for use as a SCM. Further research is in progress to investigate the wider range of concrete properties, including durability.

From an environmental perspective, a desk top study has indicated that processed CDFA would have a carbon footprint of 96.5 kg CO_2 per tonne. As a consequence, every tonne of processed CDFA used as a SCM would result in a saving of some 760kg of CO_2 together with replacing some 1.6 tonnes of limestone and shale.

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APPENDIX A1: Landfill Operating Plant: 250,000 tonnes recovered 99% yield

Machine Name	Litres per hour	Number of machines	Litres per hour total	Hours per week	Total litres per week	Total litres per annum
Dozer	25	2	50	50	2,500	130,000
Loading Shovel	20	4	80	50	4,000	208,000
Dumper	18	2	36	50	1,800	93,600
Screen Plant	15	2	30	50	1,500	78,000
Other Kit on fuel	10	1	10	50	500	26,000
Tractor Bowser	15	2	30	50	1,500	78,000
				Total litres of fuel		613,600

Diesel combustion	720	g carbon/l
	1,920	g oxygen/l
	2,640	g CO_2/l
Fly ash recovered	250,000	t

Landfill operation CO_2 = (total litres of fuel x diesel combustion CO_2 per litre) / fly ash recovered

= (613,600 x 2,640) / 250,000) = 6,480 g/t extracted

Total landfill operation CO_2 = landfill operation CO_2 grams per tonne x fly ash recovered

= 6,480 x 250,000 = 1619904000 g CO_2 = 1620 t CO_2

APPENDIX A2: Drying: 247,500 tonnes dried 80% yield

Wet ash rate	20	t/h
Moisture	20	%
Dry solids out	16	t/h

Gas requirement

Gas required 450 M^3/hr

= (450 x 1000) / 22.4 = 20089 mol/h *Note: At STP 1 mole of gas occupies 22.4 litres*

Assumed gas composition	CH_4	88.6%	($CH_4 + 2O_2 > CO_2 + 2H_2O$)	*Note: 1 mole of CO_2 per mole of CH_4*
	C_2H_6	5.3%	($2C_2H_6 + 7O_2 > 4CO_2 + 6H_2O$)	*Note: 2 moles of CO_2 per mole of C_2H_6*

CO_2 (mol/hr) = (moles of CO_2 per mole of CH_4 x CH_4 percentage x gas required)

+ (moles of CO_2 per mole of C_2H_6 x C_2H_4 percentage x gas required)

= (1 x 0.886 x 20089) + (2 x 0.053 x 20089) = 19929 mol/h

CO_2 (kg/hr) = moles CO_2 x (kg oxygen + kg carbon)

= 19929 x (0.032 + 0.012) = 877 kg/h *Note: 2 moles of oxygen = 32g, 1 mole of carbon =12g*

CO_2 (kg/t) = kg/hr CO_2 x dry solids out

= 877 / 16 = 54.8kg/t

Electricity requirement

Mills required 2 for processing 20 t/hr

Per mill:

Dryer	Motor (kWh)	Operating (kWh)
Mill	250	125
Rotary V	1.5	1.5
Screws	1.5	1.5
Fan	200	140
Total operating kWh per mill		268

Total kWh = mills required x total operating kWh per mil = 2 x 268 = 536 kWh

Total kWh on dry feed basis = total kWh / dry solids out = 536 / 16 = 33.5 kWh/t

UK electrical emission factor = 0.21233 kgCO_2/kWh

CO_2 (kg/t) = total kWh on dry feed basis x UK electrical emission factor

= 33.5 x 0.21233 = 7.11 kg/t

Total CO_2 from drying

Total CO_2 from drying = gas requirement CO_2 + electricity requirement CO_2

= 54.8 + 7.11 = 61.92 kg/t

APPENDIX A3: Processing: 199,238 tonnes processed 75% yield

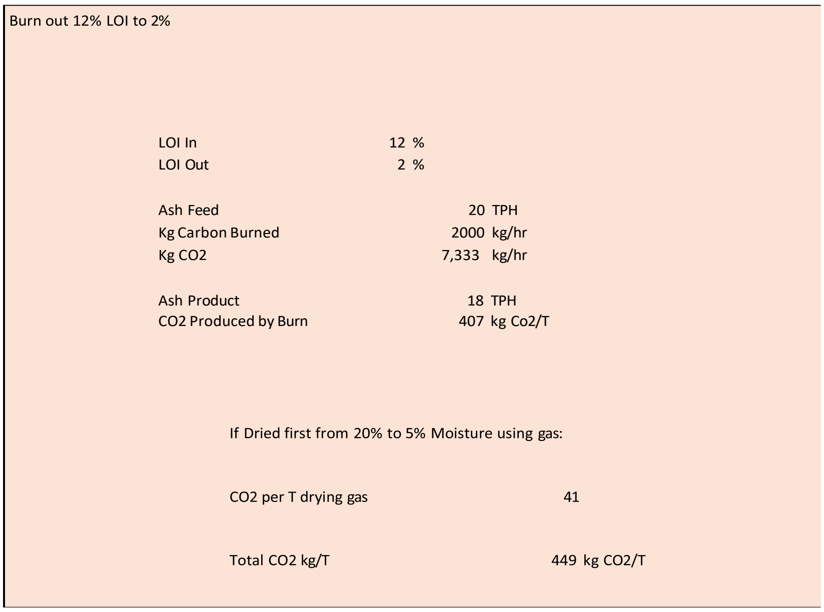
Separation plant 13.5 kWh/t

UK electrical emission factor = 0.21233 kgCO_2/kWh

CO_2 (kg/t) = (total kWh/t x UK electrical emission factor) / yield percentage

= (13.5 * 0.21233) / 0.75 = 3.82 kg/t

APPENDIX A4: Carbon Burn Out Assumptions



Burn out 12% LOI to 2%

LOI In	12	%
LOI Out	2	%
Ash Feed	20	TPH
Kg Carbon Burned	2000	kg/hr
Kg CO2	7,333	kg/hr
Ash Product	18	TPH
CO2 Produced by Burn	407	kg Co2/T

If Dried first from 20% to 5% Moisture using gas:

CO2 per T drying gas	41	
Total CO2 kg/T	449	kg CO2/T

FINAL PRODUCT SUMMARY

Process	Ash (tonnes)	Yield (%)	CO2 (tonnes)	Split	Product (kg CO2/t)
Landfill Extraction	250000	99	1620	11.23%	10.8
Drying	247500	80	12231	84.81%	81.9
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Final Product	149428		14422	100.00%	96.5

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