

Review of Existing Technologies for the carbonation of End of Life Cement Fines

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Contents

Introduction

Development of Recycling Process

- Current Industry Process
- Advanced Dry Recovery
- Isolation of End of Life Cement Fines
- Reflection on the recycling process

Carbonation

- Material Loading
- Agitation
- Dissolution of Carbon Dioxide
- Particle Dehydration

Conclusion

Referances

Page: 1

2

6

12

13

Introduction

There are a variety of hurdles for the concrete industry to surpass in order to progress from its current state to a point where the carbonation of end-of-life cement fines (EOLCF) can be achieved at industrial scale. These areas of required development can be broken into two primary groups.

-The development of recycling infrastructure to isolate EOLCF from other components present in the EOL concrete

-The carbonation of EOLCF and preparation for integration back into the cement & concrete lifecycle

This paper outlines an assessment of current technologies for the completion of the processes. Both parts of the process were considered as they are equally essential in achieving the end goal of this project which is to provide a road map to reducing carbon emissions in the industry.

Energy usage is a critical part of this process. The national grid in the UK is still heavily reliant on combustion of fossil fuels for energy generation. Therefore, every kWh of national grid electricity used comes with an intrinsic carbon cost which is considered to be around 0.435 kg CO₂/kWh^[1]. The predicted emission savings from the complete carbonation per 100g of EOLCF are 42g of CO₂ directly sequestered and 86.5g offset due to use of the carbonated material in clinker substitution^[2]. Therefore, any proposed system must use <2632kWh/ton to have any carbon-cutting effect at all.



Current Industry Process

The recycling process of construction and demolition waste (CD&W) relies heavily on the use of crushers and sieves to sort aggregate material by particle size. There are various methods for achieving this grinding, figure 1. These methods can produce a range of sizes but all produce very fine material as a by-product^[3]. This is generally low-quality material that is used for aesthetic purposes or as backfill material^{[4] [5]}.

This fine mixture of materials called 'sifting sand'^[6] or 6N fines contains a large variety of materials and impurities. A typical composition of sifting sand can be seen in Table 1.

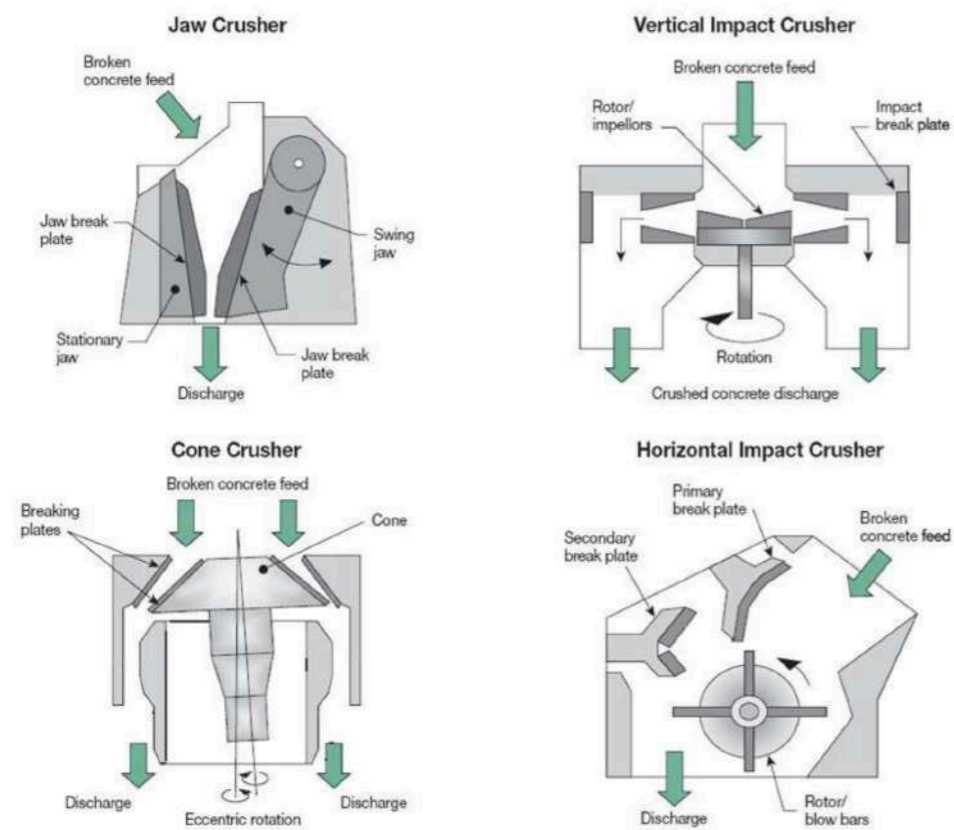


Figure 1: Material Grinding Methods^[3]

Constituent	Concentration
Fines (0-1mm)	40%
Minerals	30%
Glass	5%
Gypsum	10%
Metals	0.5%
Burnable Material	4.5%

Table 1: Composition and value of Sifting sand^[6]

Advanced Dry Recovery (ADR)

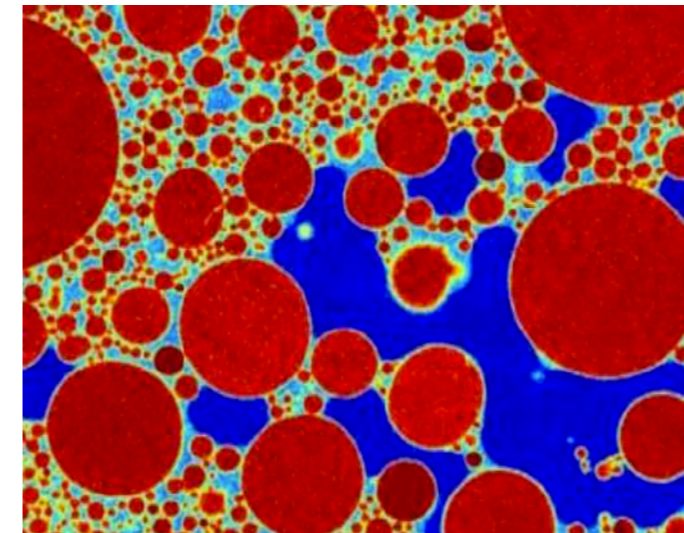


Figure 2: Image of fines interaction with water (Largest particle is 1mm)^[6]

Given the large variation in material properties of the constituent parts of sifting sand, it is difficult to extract value without further sorting. Sorting of the constituent parts is difficult due to the high moisture content present and how it affects small particles. Fines (< 1mm) interact with water by developing bonds between particles, as can be seen in Figure 2. These interparticle bonds make "Efficient classification below 8mm impossible using existing technologies"^[6]. This is mainly due to the clogging effect that the sludge-like mixture can have on sieves. The carbonation of EOLCF requires very fine particle^[7]. Given the low value of sifting sand, and the presents of small particles it is the rational choice for sourcing EOLCF for carbonation. However, to achieve this, the sifting sand must be properly sorted.

The advanced dry recovery system is a new technology that can effectively sort different sized particulates through the use of rapid acceleration. The ADR unit applies this acceleration to a stream of particles with the use of a rotating drum, Figure 3. The drum is fitted with blades perpendicular to the tangential plane of the drum. As a stream of particles is fed onto the rotary drum, they collide with the blades. This produces the rapid acceleration of particles which is necessary to break the bonds formed between particles and water molecules. The particle deceleration is defined due to air resistance and so lighter particles do not travel as far. This allows particle sizes to be separated based on the distance they travel. Any small particles that do make it as far as the fast-moving conveyor are then removed by a second sorting mechanism. The 'air-knife' is an air jet that blows lighter particles off course, separating them from the larger particles.

This technology is valuable to the project as it demonstrates that there is a feasible and cost-effective method of sorting the 'Sifting sand' produced from standard recycling processes. This is essential to achieve the end goal of EOLCF carbonation, but ADR can only separate the 2mm particles from larger particles and impurities. Further processing is required to isolate the EOLCF within this 2mm fraction.

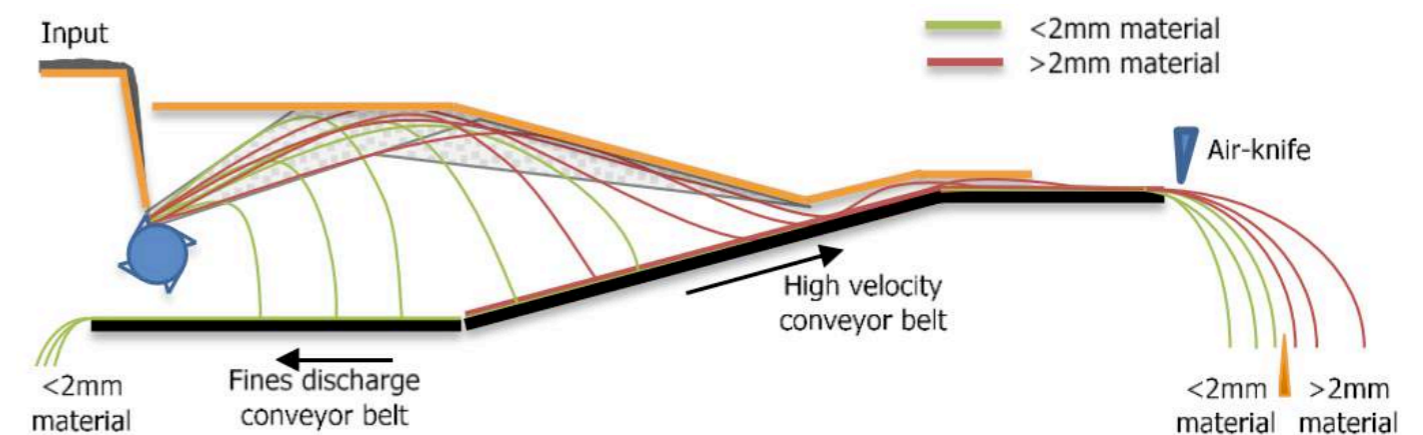


Figure 3: Design of an ADR Unit^[6]

Isolation of End-of-Life Cement Fines (EOLCF)

There is no existing scalable solution for the isolation of EOLCF. Despite this, experimental work carried out on behalf of C2CA (Concrete to Cement Aggregate Company) has demonstrated a feasible method for separation of EOLCF from other larger particles in the <2mm range, separated by ADR.

Heating-Air Classification System

The general overview of the experimental process can be seen in Figure 4. This allowed for the isolation of particles less than 0.25mm. The experimental apparatus can be seen in Figure 5. The <2mm fines removed from the ADR were fed into a tube on an incline plane. A stream of air was applied to the system from the bottom end of the pipe to separate heavy and light fractions of the fines. The pipe was heated with Bunsen burners to dry the fines. The use of natural gas combustion, in this case, seems counterproductive but the researchers concluded that "a heating step in the recycling process seems inevitable to make the process robust."^[8]

The larger particulates were collected in the hopper at the lower end of the tube, these were then crushed using a ball mill and sieved to 0.25mm. Testing at various temperature showed that the heating element weakened the bonds between sand particles and cement particles so less ball milling was required to separate the two.

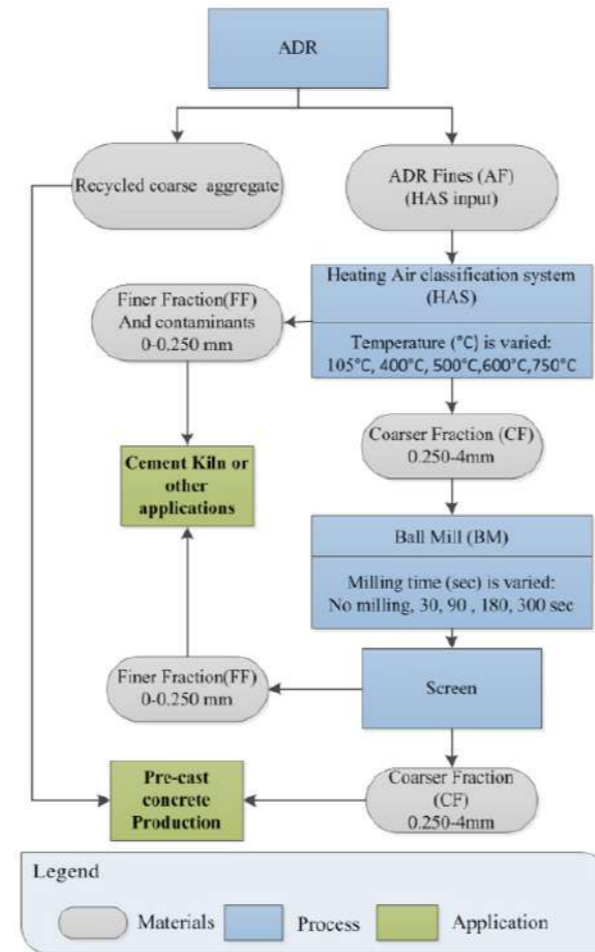


Figure 4: Flowchart of the Recycling Process^[9]

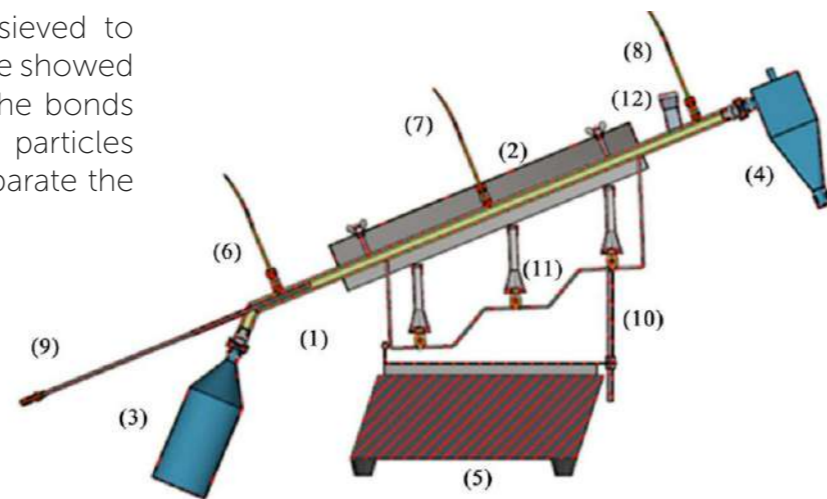


Figure 5: Heating air classification system experimental apparatus^[9]

Reflection on the Recycling Process

As outlined above, the current recycling process for concrete does not possess the infrastructure at scale to provide the material necessary for carbonation of

EOLCF. However, the description of developing technologies demonstrates that the industry recognises the area for improvement of efficiency. This shows positive growth and gives confidence to the eventual success of the project.

Carbonation Process

To achieve complete carbonation a variety of different sub processes need to be completed. The complete flow diagram for the process can be seen in figure 6. The process steps that can be seen here have a varying level of complexity. This research focused on the areas that are likely to cause technical problems or have high energy usage and omitted the simple processes such as hydration of the EOLCF.

Material Loading

Given the size of the industry, this is not a new problem in cement manufacturing and as such the development of material movement is at an advanced stage. The most commonly utilised method is the use of lifting or moving conveyors as they offer a fully automated and continuous solution. Archimedes screws are also fairly common when transporting more liquid or sludge-like material. Both of these provide fully automated low power solutions with a flat moving conveyor using approximately 1kw/ton meter^[10].

Agitation

There are a variety of ways to effectively mix the water, EOLCF and CO₂. A good example for this process can be found in the processing of wastewater treatment plants. This is an apt comparison as both processes require constant agitation and integration of gas. They will also both function on an industrial scale. Agitation methods could use either mechanical mixing methods such as propellers or use fluid pumps. Either method requires the use of electrical motors or pumps. A large scale water treatment plant used five 75kW motors and five 37kW submerged pumps to achieve full agitation of an 8015m³ plant^[11]. Assuming a processing time to achieve complete carbonation of 6 hours^[12] and a setup as mentioned above, Carbonation of a full tank would use 2250 kWh to power the motors & pumps. Assuming 90% water content by volume would lead to an approximate energy usage 4.19 kWh/ton of EOLCF and hence 1.82 kg of CO₂/ton of EOLCF carbonated. This is less than 2% of the emission that would be sequestered or offset by the process.

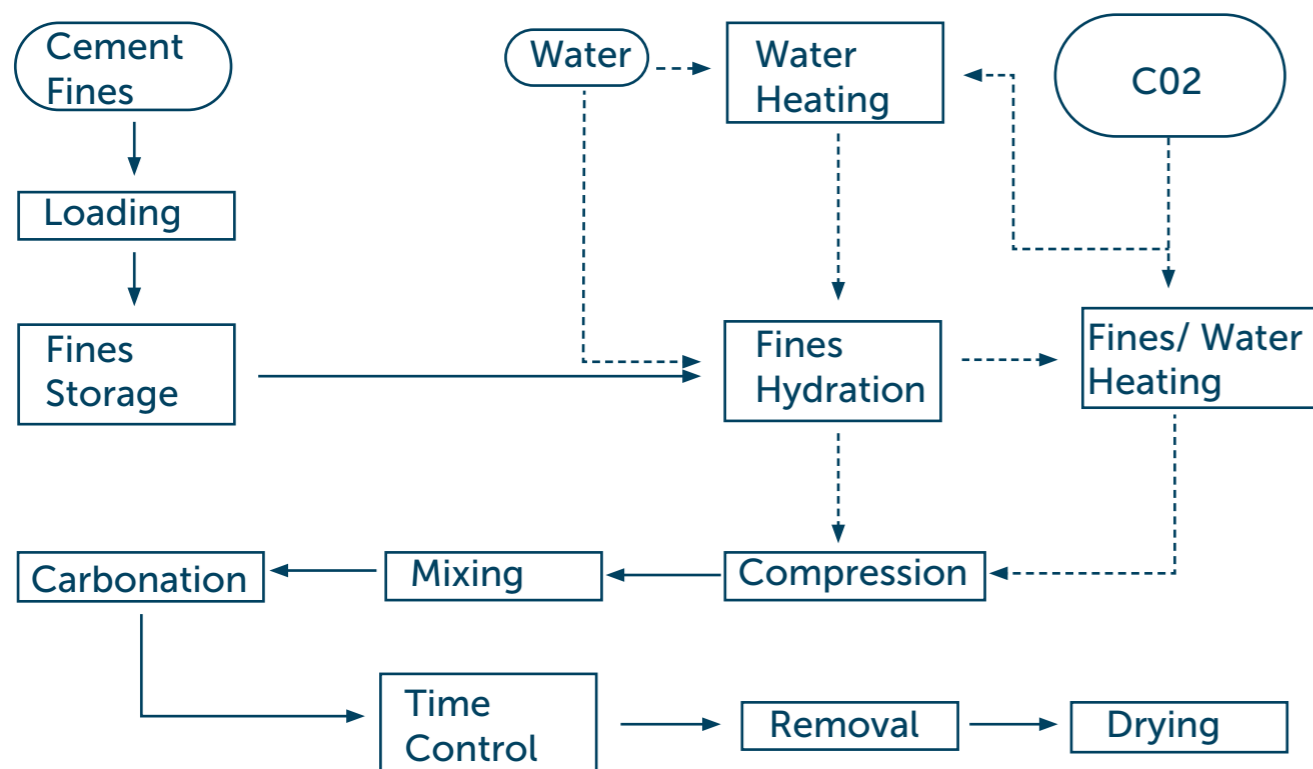


Figure 6: Carbonation Process Flow digram

Dissolution of Carbon Dioxide

A critical part of the carbonation process is the dissolution of carbon dioxide in water to form carbonic acid^[13], which reacts with the calcium present in the cement fines to form limestone. There are a variety of existing technologies for optimising dissolution of gases in water that could be transferable to this application.

Diffusion membrane

Diffusion membranes are commonly used for aeration in sewage processing plants. These can function at low pressure and offer a passive method of integration. This works by passing air through a thin tube to maximise the surface area to volume ratio. The tube wall is made of a semi-permeable membrane that allows for diffusion of air from the tube into the fluid surrounding it. This represents proof of concept at scale as the technology is commonly used at industrial sewage plants^[14].

Phanpa et al^[15] showed that a similar methodology could be used for the capture of CO₂. Experiments were completed with both hydrophilic and hydrophobic membrane materials, with the latter performing slightly better. The experimental setup can be seen in Figure 7.

These experiments were carried out using air mixed with 9000ppm of CO₂, as the test was to analyse the possibility of removing CO₂ from the atmosphere.

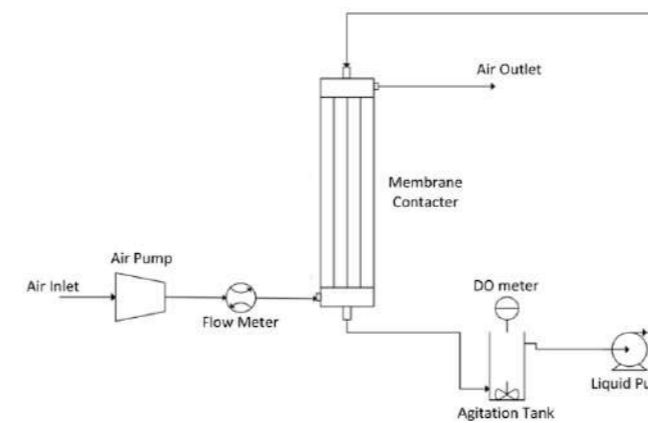


Figure 7: Experimental Setup for testing oxygen collection form air (was also tested for CO₂ experiment)^[15]

The results can be seen in Figure 8 and show the efficiency of CO₂ removal. A Sodium carbonate solution was also used to acceleration the carbon diffusion rate.

The exhaust gases from the rotary kiln contain approximately 25 mol%^[16] carbon dioxide. Therefore, it can be reasonably assumed that by using such a concentrated supply of carbon dioxide, a greater concentration gradient would lead to an increase in diffusion rates. This design approach also has the significant advantage that it does not mix any particulates or other gases present in the exhaust fumes with the EOLCF solution.

This technology could be highly applicable to the use in the carbonation system as it provides a possibility for a low energy method of integrating carbon dioxide directly into the cement fines solution. This method has the advantage that it is completely passive but would require a large quantity of membrane tubing to handle the flow rate output of exhaust gases. It is also difficult to predict what rate of CO₂ removal from the exhaust gases could be achieved without experimental data.

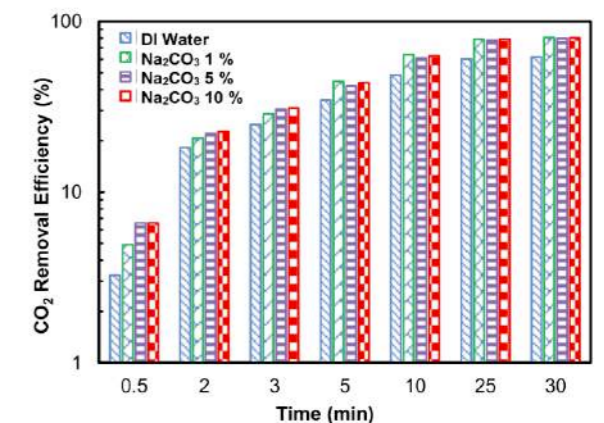


Figure 8: Rate of CO₂ removal through a semipermeable membrane^[15]



Industrial carbonation

Another method for carbonation of the EOLCF solution would be the direct integration of CO_2 into the solution. This would function similarly to a soda stream on an industrial scale.

The rate of reaction between carbonic acid and EOLCF is heavily influenced by the number of interactions between molecules, this has been shown under experimental conditions, as "generally, the higher the pressure and temperature, the higher carbonation extent" ^[2].

The relationship between CO_2 solubility with pressure and temperature can be seen in Figure 9. As outlined in the first paper of this series^[7], the optimum temperature for carbonation of EOLCF is 80 °C. At this temperature, the solubility of CO_2 will be fairly low. Therefore, to achieve a fast reaction rate, a high-pressure environment would be required. The same could be said for the diffusion membrane approach, though the low solubility could be offset by increasing the membrane piping length.

It has been demonstrated that compression of CO_2 up to 110 bar requires an energy demand of 0.146 kWh/kg CO_2 ^[16]. Using this as an approximation for compression of exhaust gases, the emissions from high compression would be 254 kg CO_2 /ton of CO_2 , enough to fully carbonate 2.38 tons of EOLCF. Under these conditions, the emissions related to compression would counteract 9.4% of the emissions saved from the carbonation process.

This shows that industrial compression is a fairly expensive process in terms of carbon emissions. Based on this, an ideal design would rely on minimal compression. This may lead to a reduction in reaction time but a balance between power usage and reaction time will need to be defined.

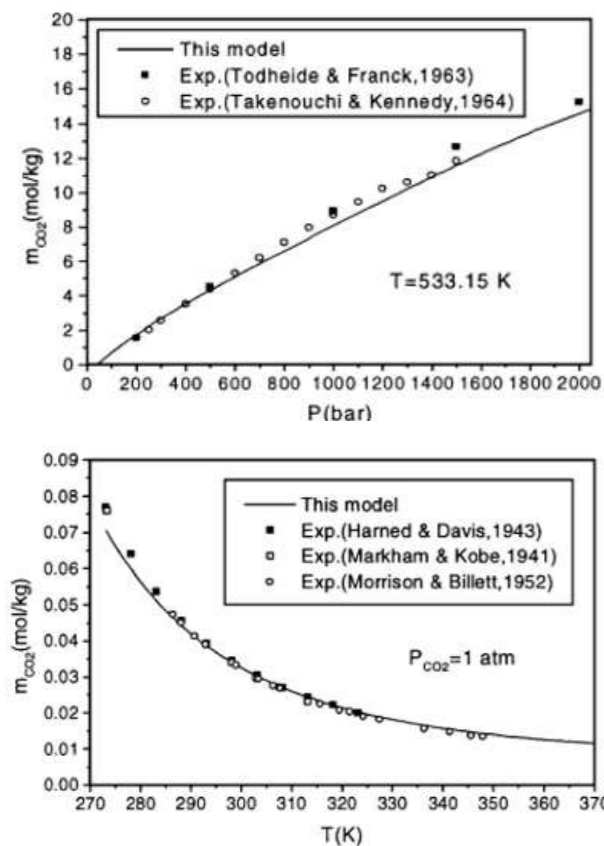


Figure 9: CO_2 solubility in pure water^[17]

Particle Dehydration

A significant part of the carbon emission reduction comes from its use as a clinker substitution material. To allow for easy integration of the fully carbonated EOLCF must be of equal moisture content to prevent binding of the material. The moisture content of clinker can vary from 6-16% after leaving the rotary kiln^[18]. To achieve complete carbonation of the EOLCF, it must be fully saturated to allow for the proper formation of carbonic acid^[12]. Hence a significant amount of water removal will be required after the carbonation process. As is the case with sifting sand, as mentioned above, the necessity for small particle sizes of EOL cement^[7] will lead to similar bonding between particles and water molecules.

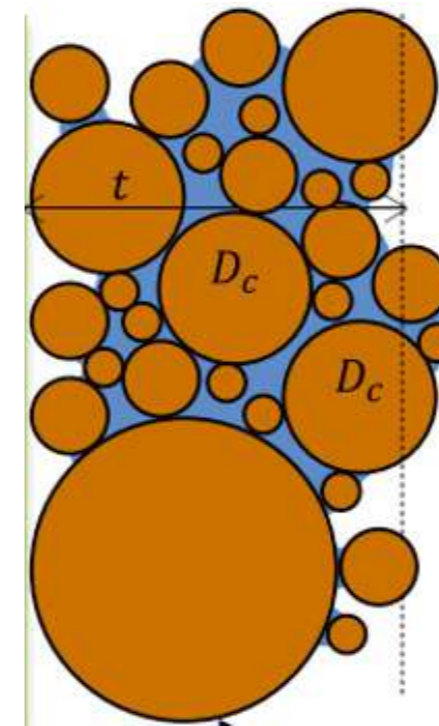


Figure 10: Interaction between small particles and water^[6]

There are a variety of available methods that could be used in this case. The critical requirements for this process are to achieve an acceptable moisture level whilst keeping energy usage as low as possible. All energy has a carbon cost and so this should be minimised wherever possible.

There are two feasible methods to achieve the required moisture content of the carbonated EOLCF, Filtration and drying.

Rotary Dryer

The rotary drier works similarly to the rotary kiln. Material is fed onto a rotating drum on an incline plane. The air in the drum is superheated by combustion of natural gas. Whilst being a very effective drying mechanism, the energy usage and carbon emissions caused by the combustion of natural gas are very high. Rotary drums produce 409 kg CO_2 / ton of dried material^[19]. This represents just under a third of all the carbon savings from the carbonation process.



Filter press

The filter press is a well-established existing technology that can be applied for a broad range of uses such as food processing, chemical manufacturing, mining and aggregate, asphalt and cement production. The system functions by pumping the slurry, in this case, saturated EOLCF, through a set of plates as can be seen in figure 11. The slurry enters through the hole in the middle of the plates and fills the cavities between plates. The system is then sealed and the cavity between plates is compressed with the use of hydraulics. This compression forces the water out of the cavity through the filter plate. When the compression is complete, the plates are separated and the dry material 'cakes' that have been formed are forced out of the cavity.

This technology seems to be an excellent fit for the carbonation system. It can achieve moisture levels as low as 6%^[20], which is ideal for mixing with clinker. The energy demand for this system is 11kWh/ton^[21] of sludge dried. The filtration system is electrically powered leading to an emissions rate of 47.85kg CO₂/ton, assuming 90% saturation. This system represents an 89% carbon saving when compared to the rotary dryer method.

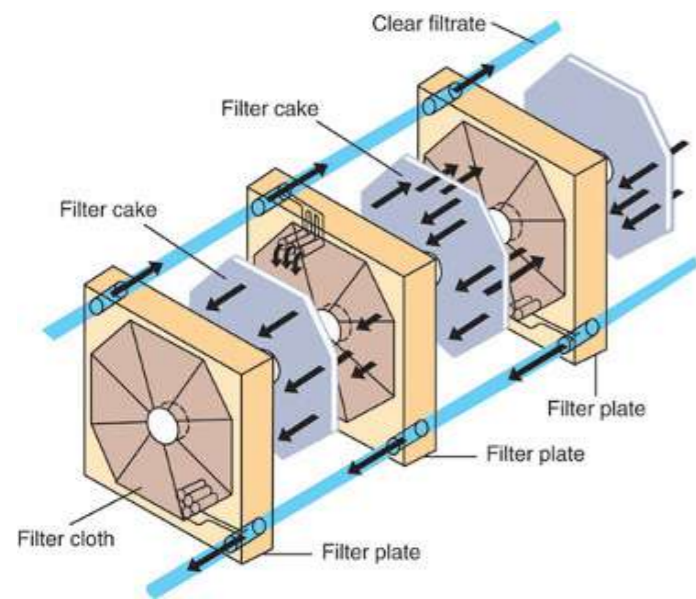
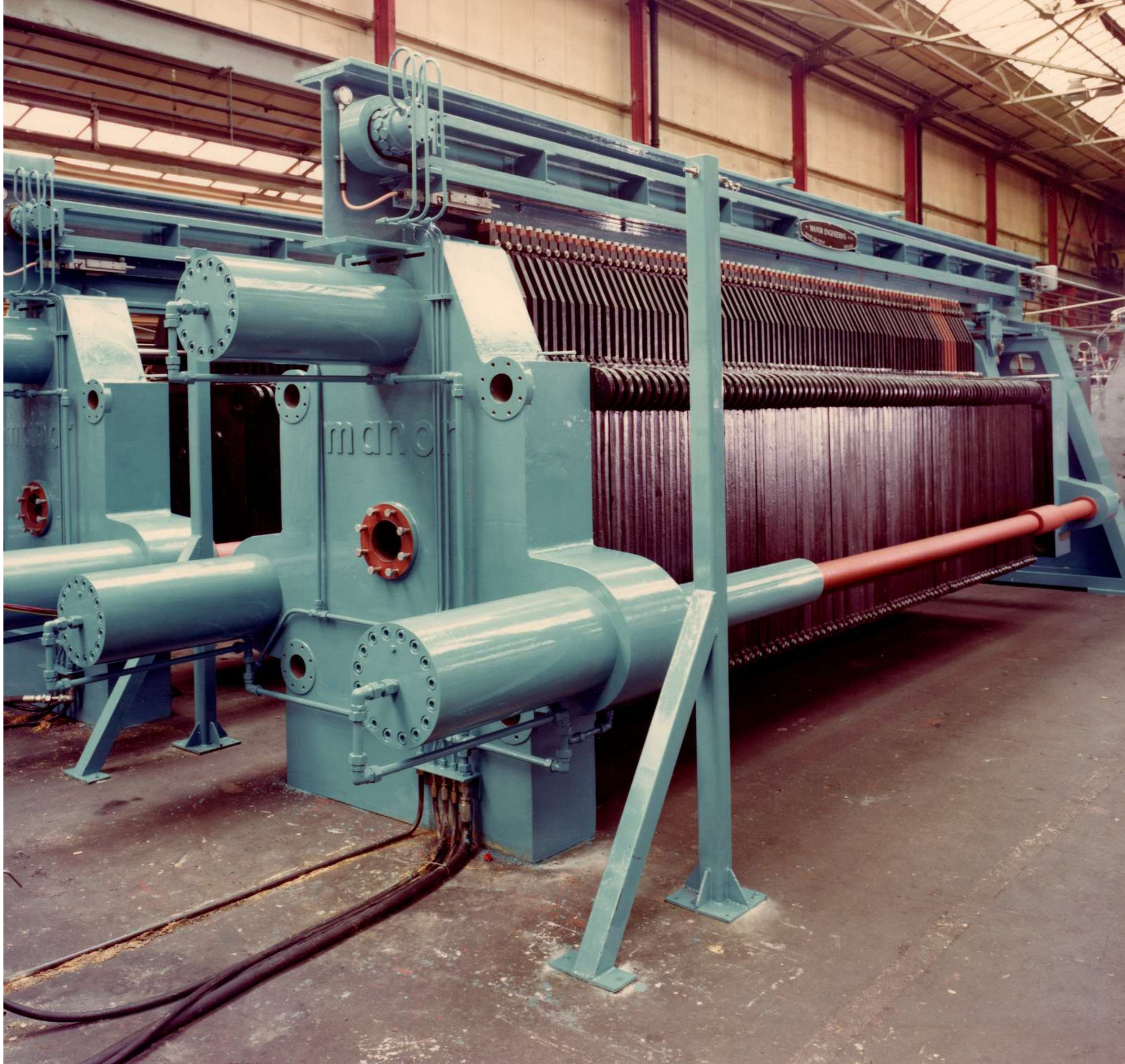


Figure 11: Filter Press Diagram



In this paper, the entire material path was considered, from the existing recycling process through to the integration of carbonated cement fines back into the life cycle of cement at the manufacturing stage. Existing technologies and theoretical methods were considered for all the critical processes between these two points.

The technologies involved in recycling and isolating EOLCF were discussed primarily to demonstrate the likelihood of the industry developing feasible methods at scale. Though further detail of this section is outside the scope of the project, it is important to understand the energy requirements to assess the feasibility of the system as a whole. It was showed that isolation of particles <2mm is achievable at scale with relatively low energy demand through the use of the Advanced Dry Recovery system. The isolation of cement particles may prove to be more difficult and energy demanding. This was based on experimental research as no existing technology has yet been developed.

The carbonation of EOLCF proved more promising as there are existing technologies that could be adapted for use in the carbonation process for all of the critical processes. The pressurised carbonation process was the most energy demanding after the rotary drier. Assuming this could be avoided through the use of diffusion membrane piping, these results are promising for the development of a low energy carbonation system.



Process	Emission kg CO2/ ton of EOLCF
Material Loading	0.435/ meter
Diffusion Membrane	Negligable
Direct Carbonation with compression	106.7
Agitation	1.82
Rotary Drier	409
Filtration	47.85

Table 1: Emissions associated with carbonation processes

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