

Postcombustion CO₂ capture with CaO. Status of the technology and next steps towards large scale demonstration

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ABSTRACT

Postcombustion CO₂ capture using CaO requires a large scale circulating fluidized bed (CFB) reactor as CO₂ absorber, operating between 600-700°C. In addition, a large scale oxy-fired CFBC must be interconnected to this reactor to allow for the decomposition of CaCO₃ formed in the carbonator. This allows for a continuous regeneration of the CaO sorbent and the production of a CO₂ rich stream suitable for final purification and compression. Despite the known limitations associated to this technology (mainly sorbent deactivation, solid attrition, and high energy requirements in the calciner) clear operating windows have been identified at which the process could be implemented in practice using the know-how on CFBC technology. Theoretical studies on the thermal integration of a carbonation-calcination loop in new and existing power plants have shown that the technology has the potential to achieve a substantial reduction (around 30%) in capture cost and energy penalties with respect to stand-alone oxy-fired systems. Since the solid materials and operating conditions in the CFB units are similar to those present in existing large scale CFBCs, the prospects for a rapid scaling up of the technology are very promising. A rapid development is taking place in recent years by demonstrating the key concepts in laboratory scale test rigs of 10s of kW. However, it is essential to move on to the next phase of pilot testing and validate the results in conditions fully comparable with those expected in large scale units. We describe here the design of a 1 MW_{th} pilot plant to capture 70-95% of the CO₂ contained in the flue gas from a 1/150 side stream emitted by an existing 50MW_e CFB power plant. The pilot is made up of two interconnected CFB reactors of 15 m height. The construction of the pilot has been initiated and is expected to enter into full operation in the first half of 2011, providing the necessary experimental results to decide on the launching of an aggressive development programme that aims to demonstrate the technology at large scale well before 2020.

INTRODUCTION

Fossil fuels account for more than 80% of the primary energy consumption worldwide and this scenario is likely to continue during next few decades. The combustion of these fuels is increasing the concentration of greenhouse gases in the atmosphere and contributing to the climate change. For this reason, the abatement of GHG derived from fossil fuel utilisation is now an urgent challenge. Between the different alternatives to stabilize GHG concentrations in the atmosphere, CO₂ capture and storage (CCS) technologies are a key option in the portfolio of solutions because they could contribute between 15 to 55% to the total mitigation effort during this century [1].

The main technical drawback of existing CO₂ capture technologies for coal based power plants is the penalty associated to the capture process. CO₂ capture technologies can be classified in two main groups attending to their state of development, near-commercial and emerging technologies. The first group includes technologies already developed at commercial scale for other processes, but that must be adapted to capture the high CO₂ flows from fossil fuel power plants and all other pollutants. The second group includes a group of technologies with a lower degree of development, but with an important potential for reducing the energy penalty and CO₂ capture costs. Between the emerging technologies, two categories can be distinguished. The first refers to processes with no analogous reactor of sufficient large scale in operation and that require a full scaling up from laboratory size and theoretical studies. The second one includes new processes that use new materials in reactors or systems already commercially established on a large scale. Calcium looping is a promising CO₂ capture technology that fits in this second category of emerging technologies, as it relies in the utilization of circulating fluidized bed reactors operating with circulating materials (mainly CaO particles and their derived products and coal ashes) and key operating conditions close to those present in large scale CFB boilers.

Calcium looping uses the reversible reaction of CO_2 with CaO to separate it from a flue gas stream in a cyclic process. Low price natural limestones and dolomites are in principle the best CaO precursor for large scale CO_2 capture in power plants. The CO_2 absorption capacity of these CaO natural sorbents is known to decay rapidly with the number of carbonation calcination cycles [2] and a large body of literature is now available trying to better understand this process and to design methods to overcome it (see most recent review by [3]). However, it is now well known [4] that there is a residual activity of the CaO particles that allows for the operation of the capture loop by using sufficiently large solid circulation rates. For those conditions where this is not possible (because attrition losses, deactivation of CaO by SO_2 , excessive heat requirements in the calciner etc), a large make up flow of fresh limestone is required. In these cases, the operation with high consumption of limestone may be allowed by the low cost of natural limestone in many locations close to power plants and the potential synergies with desulphurization units or the cement plants (making use in the clinker oven of the purge rate of deactivated CaO and coal ashes).

In this paper we briefly review the status of development of the technology from the process perspective, in order to justify our design of a 1 MW_{th} pilot plant aimed at the capture of 70-95% of the CO_2 contained in a side stream of the flue gases from the existing CFB power plant. The design of the pilot combines the knowledge acquired in recent years when testing the Ca-looping concept at laboratory scale of 10s of kW, and the industrial experience designing and operating large scale CFBs.

PROCESS DESCRIPTION AND STATUS

Figure 1 shows a concept of postcombustion capture where CO_2 is captured from the combustion flue gas of a new or an existing power plant by using CaO as sorbent in a circulating fluidized bed carbonator operating between 600-700°C. The stream of partially carbonated solids leaving the carbonator is directed to a second circulating fluidized bed where solids are calcined to regenerate the sorbent (CaO) and to release the CO_2 captured in the second CFB. In order to calcine the CaCO_3 formed in the carbonator and to produce a highly concentrated stream of CO_2 suitable for purification and compression in the calciner, coal is burned under oxy-fuel conditions at temperatures above 900 °C. Therefore, exhaust gases from the calciner contains the CO_2 captured from the flue gases of the power plant and the CO_2 resulting from the oxy-fired combustion of coal in the calciner. Other advanced concepts where heat is transferred to the calciner using a hot stream of solids circulating from an air-fired combustor have been proposed in order to avoid the penalty associated to the air separation unit [5]. However, these processes will need a longer period of time to be developed. The option of calcining the sorbent using an oxy-fired circulating fluidized bed, first proposed by Shimizu et al [6], is one of the strengths of this process and may speed up the development of calcium looping technology. This is due to the fact that the oxy-combustion CFB technology for CO_2 capture is being developed as an independent route and it is already a more mature technology in a near-commercial stage [7].

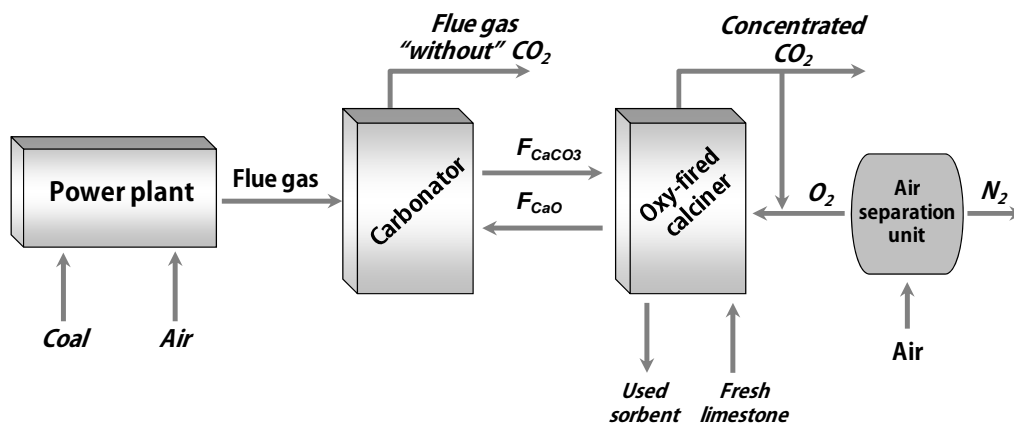


Figure 1. Schematic of a calcium looping system to capture CO_2 from an existing power plant.

A full programme to demonstrate and validate OxyCFB technology is already on-going in a collaboration among Foster Wheeler, Fundación Ciudad de la Energía (CIUDEN) and ENDESA. This programme that

counts with the support of the European Energy Program for Recovery (EEPR), includes R&D testing campaigns at CANMET Energy's 0.8 MW_{th} oxyfuel CFB pilot plant and at the CIUDEN's 30 MW_{th} OxyCFB boiler currently under design and construction in El Bierzo, Spain. The goal of this ambitious project will be a 300 MW_e commercial demonstration plant planned to start at the end of 2015 in ENDESA's Compostilla power plant, if the project is feasible. In addition and due to the higher amount of limestone to calcine in this process compared with an OxyCFB boiler, the endothermic calcination reaction can make easier the operation of the oxy-fired calciner as it may facilitate the temperature control and to reduce the requirements of recycling CO₂.

Calcium looping technology shows a low energy penalty compared with other CO₂ capture technologies [8, 9]. One of the main features of this process is the possibility of producing additionally power from the different high temperature sources involved in the system. In fact, the calciner of the process outlined in Figure 1 can be viewed as a new oxy-fired fluidized bed which increases the total installed power of the global process (power plant+power from capture process).

Some experimental works for testing calcium looping in small fluidized bed pilot plants with different configurations (10s KW) have been published in recent years. By operating a CFB pilot rig at CANMET as a bubbling fluidized bed carbonator, Abanades et al [10] demonstrated the effective capture of CO₂ from flue gases in a fluidized bed reactor with sufficient quantity of active CaO (the part of the CaO particles reacting in the fast reaction regime). Lu et al [11] reported stable capture conditions in a similar semi-continuous 75 kW pilot plant composed of an oxy-fired CFB calciner and a bubbling fluidized bed carbonator. They showed that regeneration of CaO under calcination conditions does not reduce significantly the activity of the sorbent. Charitos et al [12] performed continuous experimental tests on a dual 10 kW_{th} pilot plant composed of a FB carbonator and a CFB calciner. They reported a parametric study of the main operation variables in the bubbling fluidized bed carbonator. To achieve operation conditions closer to those expected in CFB reactors, Alonso et al [13] carried out experimental work in a 30 kW_{th} pilot plant composed of a CFB carbonator coupled with and a CFB calciner. They reported CO₂ capture efficiencies between 70 and 97 % under realistic operation conditions in the CFB carbonator reactor. A more detail comparison of the results of these testing facilities can be found elsewhere in a different communication to this conference [14].

The experimental information available at small pilot plant scale has served to establish the key variables in the carbonation process and to develop models for the carbonation reactor [15, 16]. A recent analysis of the results obtained in CFB carbonator by Rodriguez et al. [17] indicates that the overall reaction rate is limited mainly by the carbonation reaction characteristic of highly cycled solids. One of the most important variables in the capture process is the inventory of active solids in the carbonator bed. The bed of solids will be composed of a mixture of CaCO₃ and CaO (apart from CaSO₄ due to reaction between SO₂ and CaO and ashes from the coal used in the calciner). The CO₂ capture efficiency increases with the inventory of active sorbent in the carbonator bed. This is in turn dependent on the average residence time of particles in the reactor that is established by the ratio between the bed inventory and the solid circulation flow between reactors.

The quantitative information and the modelling work available from laboratory studies on the carbonator reactor has been used to design a pilot plant, following the scheme of Figure 1. However, the importance of the oxy-fuel CFB calciner, as well as the need to obtain in the pilot plant certain parameters (bed inventories, and solid circulation rates) at specific conditions, has required the application of all necessary know how on CFB technology achieved by Foster Wheeler during decades of experience in the design and construction of CFB boilers. We summarize in the following paragraphs the main design criteria, the main features of the pilot and the expected work plan.

PILOT PLANT DESIGN TO TEST THE CALCIUM LOOPIN TECHNOLOGY FOR POST-COMBUSTION CO₂ CAPTURE

As discussed in previous sections, in order progress towards the industrial application of Ca-looping technology before 2020, the next step for pilot testing at larger scale must be carried out as soon as possible. For this objective, an agreement between ENDESA (a major European utility), Foster Wheeler (a leading world manufacturer of fluidized bed combustion technology), HUNOSA (the biggest coal mining company in Spain and owner of a CFB power plant) and CSIC (Spanish Research Council) was signed in 2009. This consortium was substantially reinforced by leading R&D partners in the field in Europe (IFK of the U. Stuttgart in Germany, Lappeenranta University in Finland, Imperial College in the

UK) and in Canada (CANMET Energy), through the project “CaOling” (Development of postcombustion CO₂ capture with CaO in a large testing facility, www.caoling.eu), funded by the European Union 7th Framework Programme-FP7 (Dec 2009-Nov 2012). One of the main targets in this project is the detailed design and construction of a pilot plant of 1MW_{th}, to perform experimental testing at sufficiently large to demonstrate the performance of calcium looping as post-combustion CO₂ capture technology (operating in full continuous mode and under realistic conditions in terms of solid materials, temperature conditions, solid circulation rates and inventories, superficial gas velocities, real flue gases and oxy-combustion conditions in the calciner).

The pilot plant will be built and integrated with “La Pereda” power plant. This coal power plant consists in a circulating fluidized boiler with an installed capacity of 50 MW_e. The plant is located in Asturias (North of Spain) and is owned by HUNOSA. Figure 2 shows an aerial picture of “La Pereda” power plant and the site where the calcium looping pilot plant is being built. A bituminous coal with high ash content (ash, 36.0 %; volatile matter, 16.6 %; S, 0.7 %; HHV, 4516 kcal/kg) is used in the power plant. Typical composition of flue gas produced in “La Pereda” power plant is 5.5 %_{vol} O₂, 12.6 %_{vol} CO₂, 7.0 %_{vol} H₂O and 700 ppm SO₂.

One of the most important aims of the pilot plant is to work under operation conditions representative to those encountered on industrial CFB boilers. Following this objective, a minimum size of the facility was established according to standard design criteria of this type of reactors and the pilot plant was dimensioned to treat the flue gas corresponding to the generation of 1 MW_{th} in “La Pereda” power plant (about 1400kg/h). The core of the CO₂ capture pilot plant consists in two interconnected circulating fluidized bed (see Figure 2). Typical operation temperatures are around 650 °C in the carbonator and 920 °C in the calciner. Each reactor is equipped with a high efficiency cyclone (cut size 5 μm) and a loop seal, which can divert the stream of solids coming from the stand pipe to the same reactor (internal circulation) or to the opposite reactor. The design of the loop seal allows adjusting the solid flow between reactors. A dedicated high pressure fan is used to fluidized the loop seals and to control the solid circulation between reactors. Table 1 shows the variation range of some characteristic parameters of the pilot plant.

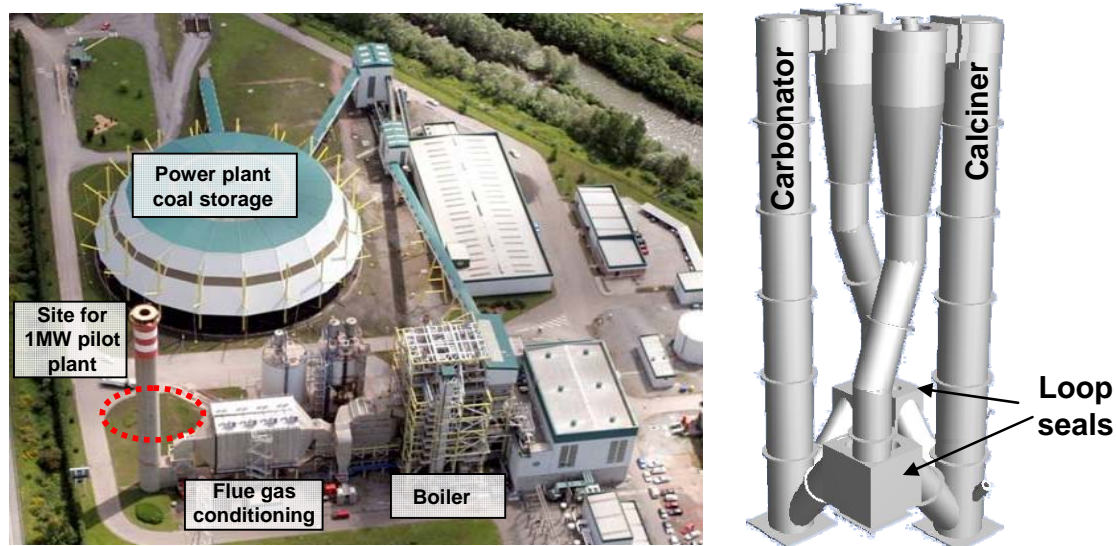


Figure 2. “La Pereda” power plant and diagram of the carbonator and calciner reactors.

The pilot plant was designed to work in an operating window where process variables can be varied as indicated in Table 1. The variables chosen to define the operation window were the flow of fresh limestone, the heat demand in the calciner and the solid circulation. Once defined the gas flow to be treated in the pilot plant and the desired capture efficiency in the carbonator, the rest of the variables involved in the process can be calculated by solving the respective mass and energy balances. The reference coal used to design the pilot plant and to solve the balances was that used in “La Pereda” power plant. Flow of fresh limestone and heat demand in the calciner were used to define the operation windows of the pilot plant, taken into account that both variables have to be minimized in a calcium looping process. The presence of SO₂ in the carbonator and ashes and sulphur in the coal fed in the calciner

affects the requirements of fresh limestone. Regarding the dimensions of the reactors, the diameter was fixed to achieve gas velocities similar to those encountered in CFB boilers (3-5 m/s). To ensure enough gas residence time and inventory of solids, the risers were designed with a height of 15 m. The maximum flow of limestone was fixed to a maximum value of 1 kgCaCO₃/kg coal fed in the calciner as these scenarios of high purge of solids can be acceptable if it is used as a feedstock in the cement industry. On the other hand, the minimum make up flow was determined by fixing the maximum heat demand in the calciner to a value of 1.7 MW. This value results in a scenario with high energy consumption in the calciner which may not be feasible from an economic point of view at large scale (the thermal power input to the calciner in large scale plants is between 0.4-0.55 of the total power input to the system). However, at this pilot plant scale it gives the possibility to increase the capacity of the carbonator to values higher than 1 MW_{th} and test the carbonation efficiency under higher gas velocities.

Table 1. Main inputs to the 1MW_{th} pilot plant.

Flue gas flow to carbonator (kg/h)	680-2300
Maximum coal flow to calciner (kg/h)	325
Maximum fresh limestone flow (kg/h)	300
Oxygen flow to calciner (kg/h)	300-600
CO ₂ flow to calciner (kg/h)	700-2250
Air flow to calciner (kg/h)	600-2500

Circulation rates expected inside the operation window can vary between 5-10 kg/m²s, which are in the typical range of CFB reactors. These solid circulations between reactors allow for effective CO₂ capture with a conservative value of calcium conversion to CaCO₃ in the carbonator. Other important aspect of the design was the heat dissipation in the carbonator, which is the addition of two terms: the flow of reacting CO₂ (exothermic carbonation reaction) and the hot solids circulating from the calciner. The first term is approximately constant once the carbonation efficiency and the flow of CO₂ to the carbonator are fixed. However, the heat transported with the solids coming from the calciner can vary greatly depending on the activity of the solids in the system and the set up to achieve a wide variety of solid circulation rates through the risers. In the most extreme case, it was calculated that the heat dissipation requirements in the carbonator can change between 0.1 and 0.7 MW_{th}.

Figure 3 shows a layout of the integration of the pilot plant with “La Pereda” power plant. A stream of flue gas from “La Pereda” will be taken after the electrostatic precipitator and will be sent to the CO₂ capture pilot plant. A fan with an average gas flow of 1400 kg/h will be used to increase the pressure of the flue gas before entering the carbonator. The maximum flow of this fan can increase the capacity of the carbonator to a value up to 1.6 MW_{th}. The gas leaving the carbonator at high temperature (approximately 650 °C) will be cooled down and returned to main flue gas stream of “La Pereda” power plant before the electrostatic precipitator.

Typical temperature of flue gas from the calciner will be higher than 900 °C. No final purification of CO₂ is planned in this pilot. Therefore the gas stream from the calciner will be mixed with the decarbonated flue gas leaving the carbonator before being cooled down. The coal feeding system was dimensioned to introduce a maximum flow rate of 325 kg/h. The system can use different types of coals or pretreated solid fuels. The fuel is discharged in a feed hopper using a bigbag handling system. From the feed hopper, coal is pneumatically transported to an intermediate bin. From this bin, coal is discharged to a common hopper where limestone and coal is mixed. A rotary feeder isolates the mixture feeding from furnace overpressure. Finally a screw feeder drives the solids into the calciner bed. As was indicated before, coal can be burned in the calciner using an oxy-fired or air-fired mode. It was decided to supply of O₂ and CO₂ by using tanks of liquefied gases as this is the more cost effective and the more flexible solution at this 1 MW_{th} scale. The liquid O₂ and CO₂ pass through several atmospheric vaporizers and an electrical conditioner. O₂ and CO₂ flows are controlled and blended in a mixer skid. This facility enables the mix of a variable O₂ and CO₂ percentage. The temperature of this flow is increased in a gas heater. An additional steam line joins to the flow previous to the calciner inlet. This steam flow comes from “La Pereda” power plant and can be controlled to simulate different compositions of the gas, increasing the operational flexibility of the plant. In order to work under air-firing mode, a fan will be installed to supply the air needed for the combustion of coal.

To work inside the limits of the operating window, the limestone feeding system was dimensioned to give a maximum mass rate of 300 kg/h. Limestone reception is discharged directly from trucks to a feed silo. From this silo, limestone is pneumatically transported to an intermediate bin before being mixed with coal. To remove the solids from the system, two pneumatically slide gates are located under the reactors. The calciner has an automatic purge removal system which consists in a water cooled screw conveyor. This screw conveyor discharges the cold ashes to a chain conveyor that leaves the material to a container. The carbonator material purge can only be made manually and after stop the plant.

A water cooling systems using double-pipe cooling tubes will be installed in the carbonator to control the temperature of the reactor. This system was designed to dissipate the heat coming with the hot stream of solids from the calciner and the heat produced during carbonation reaction. The system consists in four retractable bayonets vertical tubes that permit the control of the reactors temperature. Cooling water circulates by the bayonets and extracts the heat released by the system. A retractable system permit to move the bayonet tube and modify the heat exchange surface, regulating the total heat extracted in the reactor. This system gives a great flexibility and permits to operate the plant in a wide range of conditions.

For start up sequences, each reactor is equipped with a start-up burner fired with propane. These burners will be used to preheat the refractory of the reactors, increasing their temperature up to solid fuel ignition. The propane is supply by a liquid propane tank with all the associated equipment to gasify the fuel. A full skid of valves control the feeding process to the burners

The instrumentation of the pilot plant was defined attending to control requirements and to obtain information to analyze the experimental results (inventory of solids, solid samples, gas composition, temperature...). There will different ports along the risers, stand pipes and loop seals to measure different parameters inside the reactors and to collect solid samples. Gas composition will be analyzed at the exit of each reactor and along the risers.

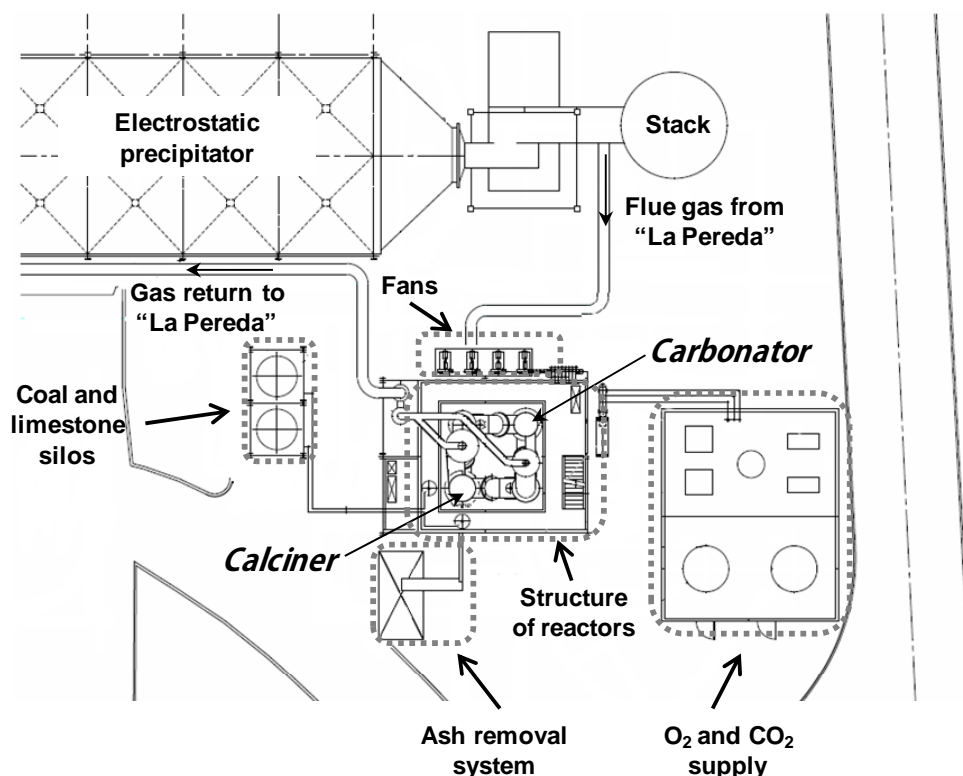


Figure 3. Layout of the integration of the 1MW_{th} pilot plant with “La Pereda” power plant.

The first experimental campaign will start in the middle of 2011, and will run for 12 months. The experimental plan will explore the impact of the main operation conditions on CO₂ capture efficiency and

other pollutant emissions. Also, due to the nature of calcium looping technology, which involves a stream of sorbent circulating between two interconnected CFB reactors, the controllability and stability of the interconnected solid circulation system is a key aspect that has to be analysed and evaluated in this experimental facility during the testing program. From the information and experience obtained during the operation of the 1 MW_{th} pilot plant, a conceptual design of a pre-industrial plant of 20-30 MW_{th} will be carried out. The construction of this medium size plant will be the next step in the scaling up route, if the results obtained in the 1MW_{th} pilot plant are successful and the validity of calcium looping under realistic CFB conditions is demonstrated.

CONCLUSIONS

Calcium looping is a promising technology for post-combustion CO₂ capture. In order to scale calcium looping at large scale, a 1MW_{th} pilot plant has been design and is being constructed close to a 50 MW_e CFB power plant in Asturias (Spain). This facility is expected to enter into full operation in the first half of 2011. The aim of this plant will be to advance in the experimental validation of this technology and to speed up the development of the calcium looping towards commercial scale. It has been possible to design this pilot using design information, equipment and material that are standard in the existing large scale CFBC power plants. This highlights the possibility for a rapid scaling up of calcium looping technology if the testing program on this pilot confirms the good results obtained so far in small lab scale units.

ACKNOWLEDGEMENTS

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