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CO₂ capture pilot test at a pressurized coal fired CHP plant

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Abstract

CO₂ capture from pressurized flue gas using a variation of the hot potassium carbonate process has been demonstrated at a pilot plant with authentic flue gas from the Värtan CHP plant in Stockholm. The plant has served as a proof of concept installation, and has confirmed that: i) the pre-treatment of the flue gas is adequate for protecting the absorbent from degradation, ii) the CO₂ capture efficiency is high (> 98%), iii) no harmful components are fed to the gas turbine, and iv) absorbent degradation is low (0,85 mole percent/month).

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1. Introduction

1.1. Background

The power producing and GHG emitting sectors, as well as the scientific community, are struggling to bring CO₂ capture and storage (CCS) technology closer to commercialization. This paper describes tests performed by Sargas AS (in collaboration with Fortum Värme AB and KTH among others) in order to verify CO₂ capture from pressurized flue gas using potassium carbonate. The site of the pilot test has been Fortum's combined heat and power plant in Stockholm that is based on the pressurized fluidized bed combustion (PFBC) technology, more specifically the ABB Carbon P200 PFBC cycle. Because of its already pressurized combustion vessel, the PFBC plant is an excellent retrofit opportunity for Sargas' technology. Another thing that makes the PFBC suitable for post combustion CO₂ capture in general is the low levels of SO_x and NO_x emissions, which is a prerequisite for low levels of solvent degradation. This initiative by Sargas is in line with, and also inspired by, the European Union's proactive stance on CCS and climate change issues in general. The initial policy documents on CCS illustrate EU's strong wish to become a forerunner and a global leader within this area [1]. In October 2008 the European Parliament's environment committee voted to back an emissions limit of 500 g CO₂/kWh on all new coal plants built

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after 2015, which is an indication that may spur the development of CCS. It was suggested to let 500 million allowances from ETS go into funding the EU Flagship Programme, which aims at realising 10-12 full-scale demonstration plants by 2015. The purpose is to prove that CCS works and is safe, and also to make sure that CCS is commercially viable for all new fossil fuel power plants by 2020.

While CO₂ capture technology in general is already commercial in niche applications such as enhanced oil recovery and natural gas sweetening, making it work also for large scale power plants, and at a reasonable cost, is still to be proven. The capital cost and the efficiency reduction that CO₂ capture incurs makes it expensive. The recent development when it comes to e.g. fuel and steel prices has added to the problem. Several projects have been put on hold lately and others have been delayed or cancelled. One recent example is the US Department of Energy's abandonment of the FutureGen project, which was announced by Secretary of Energy, Samuel Bodman in January of 2008. The deciding factor was the projected cost of the plant, which had doubled from US\$ 950 million in 2003 when the project was launched, to US\$ 1.8 billion [2]. The example shows that with higher energy prices, it gets increasingly important to develop CO₂ capture processes that have low penalties. Also, a high steel price makes it increasingly important to develop reliable, compact and integrated processes with low investment costs. The Sargas technology is interesting in this respect since the CO₂ capture is pressurised and highly integrated in the process. This gives some interesting possibilities when it comes to choice of absorbent as well as enabling a compact capture plant.

1.2. Capturing CO₂ from coal fired power plants using potassium carbonate absorption

The Sargas process for CO₂ capture has already been described in the scientific literature, both with a gas fired boiler and a coal fired boiler [3, 4, 5]. The gas fired concept has also been verified by Siemens and Alstom in 2005-2006. The demonstration unit that is described in this paper was built in order to demonstrate that the concept is also applicable on the PFBC technology and together with coal derived flue gas. This section describes earlier experience from pilot plants for CO₂ capture using real flue gas as well as scientific literature on research priorities and environmental effects regarding CO₂ capture. The material will frame the results and concluding discussion of this paper.

In literature describing demonstration of post combustion capture of CO₂ from coal-derived flue gas, the systems are often based on amine absorption, and more specifically monoethanolamine (MEA). One reason is that amine absorption systems are relatively mature in comparison with other capture technologies, and there are at least two commercial amine processes on the market. Knudsen et al. [6] evaluates the experience from the first year of operation of the MEA absorption pilot plant located at the Esbjerg coal-fired power plant in Denmark. The purpose of that particular pilot plant is to test new solvents and gain hands on experience with working on real coal-derived flue gas. Their conclusion from the first campaign, when using MEA, is that the plant could run continuously for 500 hours with an 88% CO₂ removal efficiency and a close to neutral water balance. The average steam demand for solvent regeneration was approximately 3.7 GJ/ton CO₂ and the MEA consumption due to degradation and emission losses was found to be between 1.4 and 2.4 kg/ton CO₂. In a study by Idem et al. [7], which builds on experience gained from extensive pilot tests conducted at SaskPower's Boundary Dam power station, the goal was to evaluate mixed amines by comparing MEA absorption with that of a MEA/MDEA blend. (A comparison of the performance of aqueous 5 kmol/m³ MEA with that of an aqueous 4:1 molar ratio MEA/MDEA blend of 5 kmol/m³ total amine concentration.) Blends are often said to maximize different desirable qualities of individual amines so that e.g. a primary amine like MEA (high reactivity but high regeneration energy demand) can be mixed with a tertiary amine like MDEA (higher loading capacity and lower regeneration energy demand) to create a mixture that retains the reactivity of a primary amine but has the loading capacity and low regeneration costs of tertiary amines [7]. Their conclusion is that a large reduction of the energy demand for regeneration can be realized by using a mixed MEA/MDEA solution in an industrial environment. However, the benefit is dependent on whether the chemical stability of the solvent can be maintained.

Knudsen et al. [6] as well as Idem et al. [7] bring up several problems with using amines, among them corrosion, degradation of solvent and large requirement for regeneration energy. These problems lead to high operating costs on top of the already high investment costs [8,9]. Working with mixtures seems promising but somewhat uncertain. In a questionnaire that was sent to professionals working in the area of amine-based CO₂ capture, Rao et al. [10] asked the experts to identify research priorities in order to minimize the overall cost of CO₂ avoidance when using

amine based systems. The answers showed clearly that the highest R&D objective is: i) to develop sorbents with a lower regeneration energy requirement, closely followed by ii) the need to improve the level of heat integration between the power plant and the amine system.

Potassium carbonate is commonly used for CO₂ removal in industries and processes with a high pressure, e.g. in ammonia production [11]. The Benfield process is one example of a commercial process that uses a 20 to 30% aqueous solution of potassium carbonate to capture CO₂. The selectivity and slow rates of absorption, makes the Benfield process unsuitable for coal fired power plants. However, if mixed with an amine K₂CO₃ can be used in a coal fired power plant. Oexmann et al. [9] model a coal fired power plant with CO₂ capture that uses piperazine promoted K₂CO₃ as absorbent and comes to the conclusion that the process shows energetic advantages over a reference plant that uses MEA. Instead of working with amines as promoting additives to K₂CO₃, Sargas has identified the opportunity that comes from working with PFBC-derived flue gas. The pressurized flue gas, the highly integrated process and the novel Sargas-solvent is an attempt to fulfill the research objectives described above, i.e. lower regeneration energy requirements as well as a highly integrated process that maximizes the heat integration between the power plant and the CO₂ capture system. Since the potassium carbonate is a cheap substance, the Sargas-solvent is relatively cheap compared to e.g. MEA. K₂CO₃ is also non-volatile and non-toxic, which is an advantage compared to working with MEA that has many environmental trade-offs such as human toxicity that can be ascribed to emissions of ethylene oxide during MEA production [12].

1.3. Aim & Scope of the study

The aim of this paper is to describe and provide results from the CO₂ capture pilot test conducted during 2007 and early 2008 at Fortum's CHP plant in Stockholm. The test of the Sargas power plant process was conducted in order to:

- Demonstrate adequate CO₂ capture (a minimum of 95%) from an operational coal fired power plant by treatment of a real flue-gas stream from a pressurized boiler.
- Demonstrate adequate cleaning of the flue gas to avoid settlement of particles in the flue gas stream and possible damage to the gas-turbine or other rotary equipment used to capture the energy (pressure) of the clean flue gas.
- Gather process experience and data to support final detailed design and scale-up of the Sargas flue-gas cleaning and CO₂ capture for a coal fired Sargas power plant process.

KTH has closely followed the development of the Sargas technology since 2003. KTH has had some input when it comes to designing the process and have also been involved in earlier feasibility studies [3,5]. KTH was represented in the so-called Reference Group during the demonstration and was chosen to do the scientific reporting. The documentation used for this report is foremost Sargas' own documentation of the test results, but also the report of the Norwegian Institute for Energy Technology (IFE) that was contracted to supervise and review the demo plant set-up, operation, testing procedures and results.



Figure 1: Picture of the pilot plant at Fortum's CHP Plant in Stockholm

2. System Description and experimental setup

2.1. Flue gas

The flue gas for the pilot tests was taken as 10.5 bar pressurized gas upstream of the expander. It was diverted from the gas sampling pipe of the full scale plant which makes it similar to the genuine flue gas. It was pre-cooled to about 300°C and led in a pipe with 12 mm internal diameter to the pilot plant which was located at a distance of about 20 m within the same building. In order to avoid condensation of the water vapour the tube was kept at about 300°C with insulation and electrical heating. Design gas data for the inlet of the pilot was 9.2 m³/h volume flow at 10 bar pressure and 300°C temperature (equals 47.6 STDm³/h or 60 kg/h)

2.2. Pre-treatment stages

The purpose of the pre-treatment stages is to remove dust and to absorb acidic gases (sulphur oxides, nitrogen oxides, hydrochloric acid and hydrofluoric acid) in order to minimize irreversible reactions with the absorbent. The PFBC plant at Vartan has comparatively low concentrations of acidic components due to lime addition to the fluidized bed and NO_x removal by SNCR, but additional removal is still necessary.

The first pre-treatment stage is mechanic filtration to remove remaining small particles of ash and dolomite. The average dust levels in the flue gas are about 93 mg/m³ according to long term statistics but the actual levels may vary considerably. Two parallel filters with a 2 μm metallic filter elements from Porvair are used and the removed particles are dislodged from the filter elements by periodic back-flushing with pressurized nitrogen.

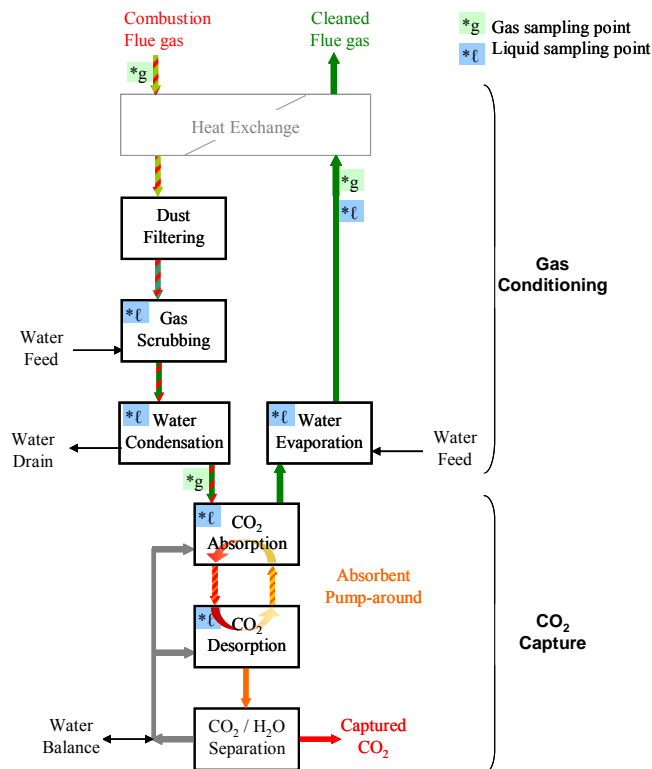


Figure 2: Functional diagram of the CO₂ capture pilot plant

The second pre-treatment stage is a co-current flow venturi scrubber, type LTB (LTB Lufttechnik Bayreuth GmbH & Co. KG) “wet throat” for injection of fine water droplets for saturation of the gas and for removal of the fine particles that have passed the metallic filter. Some absorption of soluble gases is also expected. The water consumption for the humidification was about 30 l/h and the venturi water was recycled at a flow up to 2m³/h and at a pressure drop of maximum 0.6 bar.

The third pre-treatment stage was a counter-current scrubber with a bed diameter of 0.20 m and a bed height of 2.0 m of structured packing of Mellapak 250Y. The water is recycled at a capacity of maximum 2.0 m³/h and passed a heat exchanger for maximum 25 kW of cooling. Cooling of the water circuit is used for the condensation of vapour to reach water balance for the pre-treatment. Some additional absorption of microscopic particles and entrained droplets from the venturi may also be captured. Absorption of ammonia, hydrochloric acid and nitrogen dioxide is expected to be excellent since they are easily soluble in water. Carbon monoxide (CO) and dinitrogen oxide (N₂O) have low solubility and are expected to be inert. Absorption of sulphur dioxide is dependent on pH value and may be improved by addition of alkali compounds like potassium carbonate. Absorption of all nitrogen dioxide (NO₂) from the combustor was also achieved but nitrogen oxide (NO) was not.

2.3. Capture section

The purpose of the capture section is to absorb the CO₂ by a potassium carbonate solution and to regenerate the bicarbonate solution by steam. The CO₂ absorption column is designed for 95% removal of CO₂ and the capture capacity equals 13.6 kg CO₂/h. The column has an internal diameter of 0.2 m and has 4.0 m of structured packing of Mellapak 500X. The concentration of absorbent was 165g K/kg of liquid (equivalent to 29 weight % absorbent expressed as K₂CO₃). During the absorption cycle about 30 % of the K₂CO₃ is expected to react with CO₂ (after the absorption about 60 % of the K₂CO₃ is expected to be KHCO₃ and after regeneration about 30% while the rest is K₂CO₃). Oxygen stable catalysts and corrosion inhibitors are also added.

The regeneration column has an internal diameter of 0.3 m and contains 4.5 m of structured packing, Mellapak 500X. The heat exchanger for the regeneration has a maximum capacity of 50 kW. The condenser that partially condensates water from the regeneration gas is designed for 65 kW. The regenerated absorbent is cooled (up to 20 kW) before returning to the absorption column. The cooling compensates for the reaction heat during the CO₂ absorption that tends to heat the absorbent and evaporate water. The pre-cooling of the regenerated absorbent thus ensures a self-sufficient water balance for the capture section. The pre-cooler is also used during start-up.

The outlet gas from the absorption is passed through an outlet scrubber (Di = 0.2 m, packing height 2.0 m of Mellapak 250Y, maximum water flow 2000 l/h). Its purpose is to remove droplets of absorbents to minimize losses of potassium carbonate and a demister is used to capture the smallest droplets. In a complete plant the treated gas will be reheated to about 820°C by a heat exchanger and then led to the turbo expander. If the droplets are improperly removed they will get dried and produce a dust that may give some fouling of the heat exchanger and on the expander. If exposed to higher temperature than 891°C the potassium carbonate will melt and is likely to become much more corrosive, but the turbine used in Vartan has an operating temperature of about 850°C.

2.4. Sampling and analysis

Three flue gas sampling points are installed: 1) at the inlet to the pilot plant, 2) at the inlet to the CO₂ absorber, i.e. downstream of the pre-treatment and 3) at the outlet from the pilot plant. The flue gas components CO₂, H₂O, HCl, HF, SO₂, SO₃, NO, NO₂, N₂O, NH₃ and CO were analysed by an on-line FTIR instrument during measurement campaigns. The relative accuracy was 2% and the detection limit (for wet gas), was 2.5 ppm for SO₂, 1 ppm for HCl and HF and 0.3 ppm for NH₃.

From the same sampling points physical gas samples were also taken. A sampling station with gas sampling cylinders was used for this purpose. Lab analysis of both gas and condensate collected in this way aimed at excluding the presence of specific trace impurities as well as droplets.

Six sampling points for liquid samples were installed: 1) recycle liquid from venturi scrubber, 2) from the recycle of the condensing scrubber, 3) rich absorbent from absorber, 4) lean absorbent from regenerator, 5) water from the recycle of the scrubber regenerator and 6) condensation water from cooling of cleaned flue gas.

Gas and liquid sampling (for analysis in external labs) was carried out simultaneously. Liquid sampling for operational purposes and analysis by the operators in the Värtan lab was done on a regular basis during all shifts.

2.5. Operation

The pre-treatment was installed in May-June 2007 and successfully tested in June 2007 before the summer shut-down of the CHP plant. The rest of the pilot was installed during Aug-Sept 2007. The pilot was in operation during a total time of 320 h during Oct-Dec 2007. During the operation six (four in '07 and two in '08) different sessions were run with durations of 24-104 h each. The purpose of the different sessions was to verify that removal efficiency of >95% can be obtained under different operating conditions (different flue gas flow rates, different recirculation rates and different degrees of saturation of the absorbent) and to verify good pre-treatment results for the flue gas during natural variations of the Värtan combustor(changed temperature, pressure, composition of flue gas).

The flow rate of flue gas was measured and controlled automatically after the mechanic filter.

The flow of absorbent was measured and controlled automatically after the regeneration and the temperature at the absorber inlet was adjusted by automatic control of the flow of cooling water.

The flow rate of water recirculation in scrubbers was measured and controlled automatically, and their temperatures were controlled by automatic control of the flow of cooling water.

A SCADA system registered all sensor inputs in on-line data files, operator information (remarks, chemical analysis, actions taken, attention points, deviations, shift transfer information), specific instructions and details on the ongoing campaign have been accumulated in an electronic operator logbook.

3. Test results

Table 1 contains results based on on-line gas measurements done during the “measurement campaigns”. The results proved to be consistent with the analysis of simultaneously taken liquid samples.

Table 1: Gas composition before and after pretreatment and CO₂ capture

Species	Inlet gas (by volume, wet)	After pre-treatment			After CO ₂ capture		
		Range	Average	% reduction by weight	Range	Average	% reduction by weight
CO ₂	17 vol%	17.9-17.3	17.5	2.3	0.4-0.1	0.26	98.9
H ₂ O	13 vol%	11.9-6.5	8.7	N.A.	3.6-1.9	2.7	N.A.
N ₂	67 vol%	73.2-67.0	70.6	N.A.	93.9-91.8	92.8	N.A.
O ₂	<3 vol%	<3.3-3.0	<3.2	N.A.	<4.2-4.1	4.2	N.A.
NO	17-27 ppm	17.8-10.8	15.0	28.6	15.5-8.9	12.0	56.7
NO ₂	0-7 ppm	N.D.	0	100	0	0	100
N ₂ O	37-51 ppm	47.6-36.4	40.6	1.6 ¹	66.2-41.4	54.3	-0.2 ¹
SO ₂	2-9 ppm	0-0.9	0.8	83.0	0	0	100
NH ₃	1-10 ppm	N.D.(<0.3)	0	100	0	0	100
HCl	10-35 ppm	N.D (<1)	0	100	0	0	100
HF	0-0.2 ppm	N.D (<1)	0	100	0	0	100
CO	0-9 ppm	3.8-3.7	3.8	-0.3 ²	5.7-2.3	5.2 ¹	-4.0 ¹

Carbon monoxide (CO) nitrogen monoxide (NO) and di-nitrogen oxide (N₂O) have low solubility and are expected to be inert. The confidence interval of the measurements is reflected in some “anomalies” in the balances in the table which result in a negative or very small absorption (e.g. N₂O and CO). However, when the analytical

errors are considered the absorption is interpreted as 0. During the whole time period on which the table is based, the scrubbers of the pre-treatment stages were running without external addition of alkali in order to evaluate this mode of operation. Absorption of ammonia, hydrochloric acid and nitrogen dioxide was excellent and the gas measurements showed values below the detection limit. This depends on their high solubility in water.

Absorption of sulphur dioxide is much more dependent on pH value since the absorption occurs as HSO_3^- . The removal efficiency was limited to about 80%. The explanation is probably that the pH value is too low for a more complete absorption. The acidic components in the flue gas (HCl, NO_2 and SO_2) are reacting with the basic components (dolomite dust from the fluidized bed combustion and ammonia slip from the SNCR). The SO_2 removal may be improved by addition of alkali compounds like potassium carbonate in the pre-treatment.

The Sargas concept with the pressurized CO_2 capture system as well as the Sargas Absorbent, which is a variation of the hot potassium process absorbent, allows for high capture efficiency. During the whole campaign and under all different circumstances, the percentage of CO_2 captured was systematically within the range 98–99.5 %m. Variations in load, i.e., the amount of flue gas that was fed to the capture plant varied between 86 and 108.5 %. Other variations: combustor pressure (~ 3 bar), flue gas entrance temperature (~ 100°C), CO_2 content of the flue gas (~4 %v wet), water content of the flue gas (~ 3%v wet).

The performance of a CO_2 absorption process is negatively affected by solvent degradation, which is why pre-treatment of the flue gas is important. The PFBC flue gas is in itself a good start since SO_x and NO_x levels are low to start with. The scrubber and condenser in the Sargas gas cleaning system do a good job of removing remaining impurities that could destroy the solvent. 15 % of the sulphur components escapes the cleaning system and winds up in the absorbent, however, the plan is to resolve this problem by countermeasures such as alkaline assisted scrubbing in future systems, Chlorine impurities are not found in significant levels in the absorbent. The total loss of absorbent due to impurities is 0.87 %m per month (chlorine accounts for only 0.02 %m per month). For a 100 MW full scale reference plant that has been evaluated by Fortum with capture of 640000 tonnes of CO_2 , an operational fill of 600 t absorbent containing 25 %w K_2CO_3 , and including alkaline operation of the scrubber, the absorbent loss corresponds to 0.83 kg/ton CO_2 captured.

There was an initial concern among some project partners that the gas leaving the CO_2 capture system could somehow harm the turbine. However, the samples from both the humidifier and gas condensate show that the flue gas is “turbine-ready” and is much cleaner than the initial flue gas.

The quality of the CO_2 is high and it contains no aggressive compounds from the flue gas. Before the tests had been analyzed, there was a worry that approximately 8 %v of the NO in the flue gas could end up in the CO_2 , but it was shown not to be the case. Of all gaseous impurities in the flue gas, none were found to be over the detection limit of 0.5–1 ppmv in the CO_2 .

The heat of reaction of the absorbent is lower than for most other absorbents and the heat demand for regeneration is comparatively low. However, it was not within the scope of this pilot plant campaign to verify the energy balance of the regeneration process. One reason is the simple process configuration of the pilot plant, with e.g. only a “single stage” coupling between the absorber and desorber, which makes it less energy efficient than a full scale system. Therefore, measurements and calculations on the energy balance would not render useful and representative values for extrapolation. Instead, solid operational data from existing commercial applications and results from already completed scientific studies will be used for estimating values for the energy balance of a future full-scale system. However, an after-the-fact and rather involved indirect energy balance check of the pilot plant was indeed carried out and confirmed the correctness of the estimated energy requirement of the pilot plant with 3250 ± 370 kJ/kg CO_2 (including over-proportional heat losses).

4. Concluding discussion

The pilot plant has served as a proof of concept installation, and has so far confirmed that: i) the pre-treatment of the flue gas is adequate for protecting the absorbent from degradation, ii) the CO_2 capture efficiency is high (> 98%), iii) no harmful components are fed to the gas turbine, and iv) absorbent degradation is low (0.85 mole percent/month). The pre-treatment of flue gas, which is considered the most critical part of this pilot plant campaign, has worked well with the total removal of e.g. NO_2 , NH_3 , HCl and HF. Only minor problems have been encountered during the 360 “continuous campaign” hours of operation, with e.g. some degradation of absorbent due to SO_2 slip through the scrubber when running in “acidic” mode. However, that problem is easily overcome by making the

scrubber fluid more alkaline. Compared to experience from earlier demonstrations using MEA, the absorbent loss of 0.83 kg/ton CO₂ captured, is low. Low absorbent loss in combination with low cost of absorbent will be crucial for keeping O&M costs low. The cost of MEA is currently much higher than the cost of the Sargas-solvent.

Furthermore, the treated flue gas is “turbine ready” and is much cleaner than the existing flue gas, and no traces of catalysts and corrosion inhibitors could be detected that could harm the turbine. Overall, the concept of capturing CO₂ from pressurized flue gas proved to be simple and robust; it allows a high removal rate and the use of the potassium carbonate process. The campaign was completed without compromising either safety or environmental concerns.

Some of the benefits of the Sargas concept have been pointed out in earlier studies, e.g. the compact size, reliability and maturity level of the technology [4, 5]. The drawbacks have also been discussed, among them the so called “volume penalty” that stems from the fact that the CO₂ is captured before it is expanded in the turbine [5]. While the efficiency of this concept may not be its strongest trait, the cost-advantage may come with a lower investment cost and reliability compared with those of competing options. Now it has also been confirmed that the concept works with real flue gas, with low levels of solvent degradation and negligible pollution of the treated flue gas. This is a conclusion that could not have been drawn without the tests. The most important thing that remains to be tested is the recuperator that makes up the interface between the boiler and the CO₂ capture unit. Because of its high thermal load it is considered a critical component.

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