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TNO report

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1 Public summary

In this report the latest news of post combustion capture is reported. Next to that also analysis has been made on the topics which need to be addressed to make CO2 capture more economically attractive. It is clear that a significant progress has to be made. However, looking from a retrospective point of view, it can be concluded there are already substantial improvements made.

The topics to be addressed for further research are:

Solvent development Stable solvents Low energy consuming Fast kinetics No emissions to the environment

Process intensification Compact absorbers Heat integration with the power plant Intensified strippers (eg heat integration within the stripper)

At this moment there are several potential breakthrough technologies under investigation (eg chilled ammonia). However, at this moment it is early day to report the performance of these novel capture systems.

Contents

1	Public summary	2
2	Introduction	4
3	Options to reduce CO₂ emissions from the power generation sector	7
3.1	Post-combustion capture	8
3.2	Pre-combustion capture	8
3.3	Oxy-fuel combustion	
3.4	Capture routes and technologies evaluation	9
4	CO ₂ post-combustion capture	12
4.1	Oerview	12
4.2	Post-combustion capture process challenges	14
5	CO ₂ capture activities and development lines	15
6	Research questions for CO ₂ post-combustion capture	16
7	State of the art	17
7.1	Mass transfer technology	17
7.2	Absorption systems	18
8	References	26

2 Introduction

The atmospheric concentration of greenhouse gases GHGs (e.g. carbon dioxide, methane, nitrous oxide, and chlorofluorocarbons) has increased gradually in the last century. The Intergovernmental Panel on Climate Changes (IPCC) has evaluated the size and impact of this increase. One of the conclusions is that the reasons behind the increased concentration of the greenhouse gases in the atmosphere are the human activities [1]. As a result, the global atmospheric concentration of CO_2 increased from a pre-industrial value of about 280 ppmv to 384 ppmv in 2007(see Figure 1) [2]. Moreover, the GHGs concentration is expected to increase to about 600 ppmv by 2050 if no mitigation and emissions reduction options are applied [3]. The emissions of the different greenhouse gases have been monitored and measured all around the globe. It is evident that carbon dioxide is the most important anthropogenic GHG. Its annual emissions have grown between 1970 and 2004 by about 80%, from 21 to 38 gigatonnes, and represented 77% of total GHGs emissions in 2004 (Figure 2) [3]. There is a consensus between most scientists that CO_2 emissions need to be reduced worldwide by 30 to 60 % in 2050 compared to 2000 in order to keep CO₂ concentration in the atmosphere below 450 parts per million by volume. This would keep the temperature raise between 2.4 °C and 2.8 °C compared to the pre-industrialised levels [1].



Figure 1: CO₂ global concentration [2]



Figure 2: Share of different Greenhouse gases in total global emissions in 2004 [3]

In order to achieve the Kyoto protocol targets, the European Union (EU) emphasis the necessity to reduce CO_2 emissions by developed countries by 30% in 2020 compared to 1990 levels [3] and [4]. In addition, the EU is committed to achieve a 20% reduction of its greenhouse gas emissions by 2020 compared to 1990 [5]. To reach this ambitious goal, the focus is on the power generation sector. The fossil fuels that are used for electricity generation are responsible for 41% of the global CO_2 emission in 2004 [6] (see Figure 3). In addition, there is attention towards the transport sector, which is the second largest CO_2 emitter, for reducing CO_2 emissions (e.g. by the development of more efficient engines and by switching to more environmental friendly fuel like hydrogen).



Figure 3: The global CO₂ emissions shares per sector 2004 [6]

Currently, fossil fuels provide around 80% of the world's total energy demand. Coal is playing a major role as the main source of electrical power (38% of the total electricity generation) [7]. The large dependency on fossil fuels makes it difficult to switch to other energy sources. Moreover, the world energy demand is expected to increase by over 50% between now and 2030. The prognoses indicate that fossil fuel will still be the main and most effective source of energy (see Figure 4) [8].



Figure 4: Fuel shares in global primary energy demand. *Other includes amongst others solar and wind energy* [8]

3 Options to reduce CO₂ emissions from the power generation sector

A reduction of CO_2 emission to the atmosphere from the power generation sector (main CO_2 emitters) would require a combination of several solutions (see Figure 5) [1] and [3]:

- Reduction of energy consumption
- Improve power supply and distribution efficiency
- Massive switching from coal to natural gas
- Widespread use of renewable or nuclear energy





Figure 5: Option to reduce CO₂ emissions [1]

CCS is a promising method considering the ever-increasing worldwide energy demand and the possibility of retrofitting existing plants with capture, transport, and storage of CO_2 . The captured CO_2 can be used for enhanced oil recovery, in the chemical and food industries, or can be stored underground instead of being emitted to the atmosphere. There are three basic systems for capturing the emitted CO_2 from the power sector, which are shown in simplified form in Figure 6 [1].



Figure 6: Overview of CO2 capture systems for the power sector [1]

3.1 Post-combustion capture

Capture of CO_2 from flue gases produced by combustion of fossil fuels is referred to as post-combustion capture. Post-combustion capture typically uses a solvent to capture the CO_2 from the flue gases after the combustion process. The main line of research is on the development of reactive solvents for the capture of CO_2 . Reactive solvents are the preferred choice due to the low partial pressure of CO_2 (between 3-15 kPa) in the flue gas with nitrogen to be considered as the main component (see Table 1) [10]. Nevertheless, other techniques, such as membranes, adsorption) are under development [9], [11].

3.2 Pre-combustion capture

In this concept, a fuel is reacted with air or oxygen and/or steam to give a syngas (mixture of carbon monoxide and hydrogen). This process is known as the gasification step. In the second step, the carbon monoxide is reacted with steam in a catalytic reactor (water-gas shift reactor) to produce CO_2 and more hydrogen. A physical or chemical absorption process then separates the highly concentrated CO_2 from the hydrogen (see Table 1). The hydrogen rich stream can be used as a fuel in a gas turbine combined cycle plant. Another option is to distribute the hydrogen for use in fuel cells or in the future to provide vehicle fuel [1], [10].

3.3 Oxy-fuel combustion

In oxy-fuel combustion, nearly pure oxygen mixed with recycled flue gas is used for combustion instead of air. This will result in a flue gas that is mainly CO_2 saturated with water vapour [12], [13]. Having water vapour as the main component in the CO_2 stream makes it possible to purify and store CO_2 with less downstream processing (see Table 1). If fuel is burnt in pure oxygen, the flame temperature is excessively high. However, by recycling the CO_2 rich flue gas the flame temperature will be similar to that of a normal air-blown combustor. Oxygen is usually produced by cryogenic air separation. Novel techniques with lower energy consumption and cost are being developed [1], [10].

3.4 Capture routes and technologies evaluation

Significant effort is directed towards the comparison of the three different technological routes for the CO_2 capture from fossil fuel power plants [14]-[17]. The majority of these studies came out with the conclusion that the net efficiency of the power plant, the overall cost of electricity, and cost of CO_2 avoided for the different technologies are almost comparable. In Figure 7, the efficiencies of the power plant reference cases without capture are compared with the overall power plant net efficiency after implementing the capture process. It is clear that the overall efficiency of the three CO_2 capture routes (post, pre and oxy-fuel combustions) are very similar. This supports the conclusion that the selection of one of the capture routes based on the overall efficiency is not sufficient.



Figure 7: The efficiency for different power plant technologies before and after adding the capture process [14]. *PCC: Pulverised coal power plant (with and without post-combustion capture), IGCC: integrated gasification combined cycle(pre-combustion capture), CCS: CO₂ capture, transport and storage, Oxyf: Oxy-fuel combustion power plant and LHV: low heating value.*

By evaluating the effect of the different CO_2 capture routes on the overall estimated cost of electricity, it was found that the difference between these different routes using the same type of fuel is very limited and could not be used to select a preferred capture route, see Figure 8.



Figure 8: Estimated cost of electricity in 2020 after applying different CO₂ capture routes [18]

However, the advantage of one technology route over the others could be related to the technology availability, the level of maturity of the technology, the possibility of capture process retrofitting to the existing power plants, the experience, and repetition in commercial and large-scale applications and the most important factor is the period needed for the technology implementation.

In this thesis, the focus is on CO_2 post-combustion capture route. Post combustion capture has been selected because it has the following advantages over the other technologies [19]:

- The possibility of add-on to existing power plants (retrofit possibility).
- Capture technologies are considered available and the solvent technologies are proven on a smaller scale.
- Capture readiness makes the post-combustion capture relatively easy to incorporate into power plant tackling issue with infrastructure inertia.
- It has more operational flexibility in switching between capture no capture options.
- Learning by doing will lead to cost reductions similar to experience with SO₂ capture process development.

Currently, a wide range of technologies for separation and capture of CO_2 from gas streams exist. They are based on different physical and chemical processes including absorption, adsorption, membranes and cryogenics (see Figure 9) [9], [11]. The choice of a suitable technology depends on the characteristics of the CO_2 -gas streams, which depend mainly on the power plant. As it can be seen from Table 1, for the different CO_2 capture routes different types of gases need to be separated. In addition, the pressure and the CO_2 content in the gas streams, which are important parameters in selecting the most suited technology for CO_2 removal, are different from one route to another.



Figure 9: Different technological routes for CO₂ capture [11]

Table 1: CO ₂ capture routes	classifications
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	Post-combustion (flue gas)	Pre-combustion (shifted syngas)	Oxy-fuel combustion (exhaust)
Gases to separate	CO ₂ /N ₂	CO ₂ /H ₂	O ₂ /N ₂
p (bar)	~1	10-80	~1
[CO ₂] (%)	3-15	20-40	75-95

In case of post-combustion, the CO_2 is diluted and available at low pressure that makes chemical absorption one of the most efficient options for CO_2 removal. However, in the pre-combustion route, the CO_2 has higher partial pressure that makes physical absorption or adsorption a good technology candidate for CO_2 removal. The relevance and potential of these separation technologies to CO_2 capture is subject of research and development efforts worldwide.

4 CO₂ post-combustion capture

Carbon dioxide post-combustion capture is considered one of the most mature capture technologies, since there is a good experience and reputation of this technology within many industrial applications [11].

4.1 Oerview

Rao and Rubin 2002 [11] shows that for many reasons amine based CO_2 absorption systems are the most suitable for combustion based power plants: for example, they can be used for dilute systems and low CO_2 concentrations, the technology is commercially available, it is easy to use and can be retrofitted to existing power plants. Typically, absorption processes are based on a thermally regenerable solvent, which have a strong affinity for CO_2 . The process thus requires thermal energy for the solvent regeneration. The benchmark absorption process is based on an aqueous solution with 30% by weight Monoethanolamine (MEA) as the active ingredient. This amine-based process is considered the state-of-the-art technology [20].

Fluor Daniel markets the 30% aqueous MEA solution based process under the name Econamine FGSM [20] and [24]. The solution also contains proprietary additives to prevent corrosion and solvent degradation. The presence of these additives is expected to have a negligible effect on the major performance parameters. Fluor Daniel has improved its process performance by various means, including a modification of the solvents and process under the name Econamine FG PlusSM. However, none of these novel improvements have been validated on a large-scale process. Some of the proposed improvements are based on modelling studies and lab-scale development. It is acknowledged that the solvent developments for post-combustion CO_2 capture are accelerating with the expectation of future CO_2 emissions constraints. Apart from Mitsubishi Heavy Industries marketing their KS-1 solvent [25], [26] and [27], also the CBI-Lummus [21], Cansolv and Praxair are amongst the new suppliers.

4.1.1 Process short description

The general process flow diagram for amine absorption is shown in



Figure 10. This is the typical conventional MEA-CO₂ capture flow sheet, which has been described and discussed in a large number of commercial, industrial and research activities [9], [11], [19]-[24] and [28]. The underlying fundamental principle is the

exothermic, reversible reaction between a weak acid (e.g. CO_2) and a weak base (e.g. MEA) to form a soluble salt. The inlet gas is contacted counter-currently with lean solvent in the absorber. The acid gases are preferentially absorbed by the solution. The solution, enriched with CO_2 , is pre-heated before entering the stripper where, through the addition of heat, the reaction is reversed. The lean solvent leaves the stripper and is lowered in temperature by exchanging heat with the rich solvent. The lean solvent is recycled back to the absorber. From the top of the stripper, a high-purity (dry-basis) CO_2 is produced. For the regeneration of the rich solvent large quantities of heat is required. An important aspect is the source of the heat and electricity. One approach is to produce the required heat and electrical power using auxiliary equipment. The other alternative is to extract the required heat from the existing power plant.



Figure 10: General chemical absorption CO2 capture process flow sheet

4.1.2 CO₂-Monoethanolamine chemical reactions and thermodynamics

Since the first applications of this system were in early in 1960's, a large number of studies on the reaction mechanism of CO_2 -H₂O-MEA system have been reported [29]-[36]. As a general conclusion of these studies, the absorption of CO_2 by aqueous MEA solution is based on a complex system of parallel and consecutive reactions in liquid phase. With the overall forward reaction between CO_2 and MEA has usually been represented as [34]:

$$\begin{split} &CO_2 + HO(CH_2)_2 NH_2 \leftrightarrow HO(CH_2)_2 NHCOOH \\ &HO(CH_2)_2 NHCOOH + HO(CH_2)_2 NH_2 \leftrightarrow HO(CH_2)_2 NHCOO^- + HO(CH_2)_2 NH_3^+ \end{split}$$

The first step is second-order and rate determining. The second step is considered an instantaneous reaction. However, this scheme is a substantial simplification for the reaction mechanism that actually occurs.

The CO_2 -H₂O-MEA system is considered non-ideal, which could be described in the gas phase using the Soave-Redlish-Kwong equation of state [36]. For the liquid phase, electrolyte-NRTL model is considered as a good representation of this system. The required data and parameters for this system have been developed and presented by Austgen 1989 [37].

In addition, of the main physical properties of this system (density, viscosity, thermal conductivity, and specific heat) have been measured as a function of MEA concentration and operating temperature by Cheng 1996 [38]. Moreover, the solubility and the diffusivity of CO_2 into the aqueous MEA solution have been measured and estimated by different studies, which are reviewed and summarized by Versteeg 1996 [31].

4.2 Post-combustion capture process challenges

Several researchers have studied and evaluated the MEA absorption process [11], [14], [20]-[24], [28] and [39]. Most of their conclusions focused on reducing the thermal energy requirement to reduce the overall process expenses. The literature results show different estimations for the regeneration energy requirement for the MEA process. Chapel et al. 1999 estimated the Econamine FG^{SM} energy requirement of 4.2 GJ/tonne CO_2 , which is around 36% of the overall operating cost [20]. Singh et al. 2003 [23] found by modelling the MEA process for 400 MWe coal fired power plant a specific thermal energy requirement of 176 kJ/mol CO_2 (4 GJ/tonne CO_2). Recently, Fluor claims a process development based on the MEA technology leading to a significant reduction of the energy requirement down to around 2.9 GJ/tonne CO_2 . The improvements are based on process integration and solvent improvements [40].

In conclusion, the high-energy requirement makes the capture process energy-intensive and costly. Therefore, it is important to study the conventional MEA process to have a better understanding of the process to work on reducing the energy requirement. Moreover, the general conclusion of all the studies is that by applying the current stateof-art capture process the overall power plant net efficiency will decrease with 8 to 12 percentage points ([1] and [41]). This means, for the conventional hard coal power plant that the overall electricity output will be decreased by almost 20 %. Furthermore, the CO_2 capture overall economic evaluation shows that the cost of CO_2 capture is currently very high. This implies that for realizing the potential of CCS there is a strong need for breakthrough technology.

As a result of the different CCS projects, evaluation and studies, a general conclusion can be made that the current CO_2 capture technologies have the following disadvantages:

- Significant efficiency reduction
- Increase of the power generation costs
- Huge capture plant components and equipment, which requires further development and modification
- Lack of experience in CO₂ capture process at a full power plant scale
- No global political and social decision to apply the technology and to subsides it
- Very limited public awareness of the importance of CCS technology as one of the solutions to reduce the global warming problem

5 CO₂ capture activities and development lines

The importance of carbon dioxide capture, transport, and storage (CCS) for the reduction of the overall CO_2 emission can be considered as a fact. In addition, the understanding of the challenges facing the large-scale CCS application, encourage different governments, nongovernmental organizations, research institutes, universities and many other commercial companies to invest in the CCS chain to make it ready for application in the near future.

The CCS research and development takes a significant funding and attention from all related parties. Different projects and activities, which cover most of the aspects in the CCS chain, have been started since the early 1990's.

The focus of the international institutes and organizations like the intergovernmental panel on climate change (IPCC) and the international energy association (IEA) is on the overall CCS chain definition, evaluation, legislation, international cooperation, and activities organization. However, many technical oriented projects have been carried out or planned for the future with EU or national funds. Figure 11 shows an overview of the completed, ongoing or planned CCS projects. The focus in these projects is varied from one project to another. Part of these projects focus on the development of one capture route like the European CESAR project, which focuses on CO_2 post-combustion capture. On the other hand, other projects evaluate the complete carbon capture, transport and storage chain like in the Dutch CO_2 capture and storage programme (CATO project).



Figure 11: CCS projects and activities [42] - [45] Projects with italic font are planned for the future

6 Research questions for CO₂ post-combustion capture

As has been discussed in the previous sections, CO_2 post-combustion capture is becoming an important option for CO_2 emission reduction. However, applying this technology on large-scale will not be a reality before tackling different legal, economic, social, political and technical challenges related to the current technologies. There are many questions, which need to be answered before applying large-scale CO_2 postcombustion capture:

- 1. How to reduce the additional power consumption, which results from applying the capture process?
- 2. How to reduce the overall footprint of the capture process and to make the equipments more compact?
- 3. What are the main parameters and factors that contribute in the overall cost of the capture process and how to reduce this cost?
- 4. What are the main technological areas of focus for the future development to achieve the CO_2 emission reduction with an acceptable cost?
- 5. Is there still a room for improvement on the current state-of-art CO_2 post-combustion capture process?
- 6. How reliable the capture process techno-economic evaluation tools that have been used for process evaluation?
- 7. How the CO₂ capture processes can be made reliable in power generation environment?
- 8. How to encourage the decision makers to adopt the CCS development politically and economically?
- 9. What are the methods and routes that should been followed to increase public awareness?

Most of these questions and many more need to be solved in order to apply the capture technology for large-scale applications and achieve the targeted CO_2 emission reduction.

7 State of the art

At this moment there are several vendors working on the development of post combustion capture. In this chapter a short overviews of some of the more important vendors of technology are given.

7.1 Mass transfer technology

Equipment for gas absorption processes are well known in the industry. Especially, the chemical industrial sector have a vast amount of experience for this type of operation. Companies like Sulzer have been designing and erecting this kind of equipment for decades (see Figure 16). The typical sizes which are now available are on the order of a diameter between 0.3 and 16 meter, with a tower height of 2 to 80 meter.



Figure 12: mass transfer equipment¹

The focus is now among the venders of mass transfer equipment to intensify the process by enhancing and further optimisiation process and equipment performance, leading to savings on CAPEX and OPEX. An example is the continuous improvement of packing material. The main aim is to have packing material with low pressure drop (OPEX reduction) but with high interfacial area (CAPEX reduction). Trends are to examine non-metallic packings and absorber towers. For instance concrete constructions can lead to significant cost reduction (up to 40%). However, robustness over a time period of 20 to 30 years needs to be established.

^{17 / 29}

¹ www.sulzer.com

7.2 Absorption systems

7.2.1 $Alstom^2$

Alstom decided a decade ago to invest in technology for amongst others post combustion capture. Three routes were selected and are still continued:

- chilled ammonia
- enhance amines (together with DOW Chemicals)
- antisublimation

In Figure 17, the vision of Alstom is depicted. The aim is clearly to reduce OPEX by reducing energy costs from 4 MJ/kg to 2 MJ/kg. This should lead to a significant reduction in cost of CO2 captured. Alstom thinks it is possible to reach a goal of 15 to



Figure 17: Alstom vision on post combustion capture

7.2.1.1 chilled ammonia

In the overview reports in the CATO project (eg written by TNO and KEMA) chilled ammonia has been discussed. In Figure 18, the process is schematic depicted. The principle for this process is based on:

- Ammonia (NH3) reacts with CO2 and water. It forms ammonia carbonate or bicarbonate
- Moderately raising the temperatures reverses the above reactions releasing CO2

² www.alstom.com

This technology seems to be very promising. However, major challenges are the volatility of ammonia, the low reaction kinetics and the slurry handling. The basic reaction are given below:

$CO_2 + (NH_4)_2CO_3 + H_2O \rightarrow 2NH_4HCO_3 (absorption)$ 2NH_4HCO_3 + heat \rightarrow (NH_4)_2CO_3 + H_2O + CO_2 (desorption)

It is of importance to note that there are more competing reactions during absorption, which can lead to higher energy consumption during regeneration.

The advantages are the claimed lower energy consumption and the desorption at higher pressure leading to lower cost for compression. Alstom has together with the American Electric Power (AEP) designed and build a pilot plant at AEP's Mountaineer Plant, located in New Haven, on a 30 MWth slipstream. In total 100 ktons of CO2 will be captured annually. The second phase would be a 200 MW demonstration on a 450 MW unit at the Northeastern Station, located in Oologah (USA).



Figure 18: schematic design chilled ammonia plant



In Figure 19 the envisaged timelines are given for this technology.

Figure 19: timeline chilled ammonia

At this moment there is no news available in the open literature about the performance of the pilot plants. The data is expected reported in this year.

7.2.1.2 Other techniques

No real data is available in public literature. However, it is expected that the development of the enhanced amines should lead to a mixture of two different type of amines. This lead to a predominantly carbonate forming system enhanced by a carbamate forming promoter.

7.2.2 Fluor

Fluor is a well know company involved in amongst others technology development and building of installations. The Fluor process Econamine has been discussed prior in this report. Here a short factsheet is given.

- World-wide licenses to date 24 plants
- Plants/Licenses on order: 10
- CO2 concentrations from 3% to 20% v/v
- O2 Concentrations from 1 to 15% v/v
- Only process that has 14 years commercial operation recovering
- CO2 from a gas turbine exhaust (13 to 15% v/v O2)
- Demonstrated on heavy fuel-oil boiler
- Several installed on boilers and steam reformers in Ammonia
- and Methanol plants

The largest plant is located in Bellingham (USA). This plant has the capacity of treating 330 tons/day (40MW). This is also the largest absorber (6,1 m diameter). Here below some facts are given:

- CO2 Concentration: 3.5% v/v
- O2 Concentration: 13 to 14% v/v
- 100% air cooled, zero water import or export
- Product Usage: Food-grade CO2
- Status: Continuous operation since 1991 with a 98.5% on-stream factor in 2004.

Here below some pictures are given for this installation.



Figure 20: Bellingham location

Compared with a standard flowsheet the econamine process has the following modifications:

- Enhanced solvent formulation
- Absorber Intercooling
- Lean vapor compression configuration
- Advanced reclaiming technologies
- Heat integration with the power plant

Next to that Fluor is focused on strategies to minimize the number of trains. One methodology is to enlarge the diameter of the absorber to 20 m. This would lead to a 1000 MW coal based power plant a requirement of 2 absorber trains.

7.2.3 Mitsubishi

Next to Fluor Mitsubishi is the other vendor of the so called proven technology. Based on the solvents KS-1 and KS-2 several small size plants have been erected. The solvents which have been used are based on the same ideas as described at the solvent development section of Alstom. In several CATO-reports this technology is discussed.



Figure 21a: This plant employs a chemical absorption process to separate 0.2 MtCO₂ per year from the flue gas stream of a gas-fired power plant for urea production (Malaysia, Courtesy of Mitsubishi Heavy Industries). (b) CO₂ precombustion capture at a coal gasification plant in North Dakota, USA. This plant employs a physical solvent

process to separate 3.3 $MtCO_2$ per year from a gas stream to produce synthetic natural gas.

7.2.4 Siemens

Siemens is currently working on post combustion using an amino acid based system. The solvent will be tested in a pilot plant connected to a powerplant of E.ON in Germany. The use of a salt of an amino acid should lead to a stable, low energy consuming process. The main advantage is the low amount of chemicals consumed due to chemical and thermal stability of amino acids. The system of Siemens is comparable to the development at TNO (CORAL family of absorption solvents).

In Figure 22 a view is given how a post combustion plant looks like according to Siemens.



Figure 22: PCC according to Siemens

7.2.5 HTC^3

HTC is a Canadian firm located at Regina. This firm has connection with the International Test Centre . They have developed an amine based process. This process should be low energy consuming. In the figure below some typical pictures are given.

³ www.htcenergy.com



Figure 23: modular design of a PCC unit



Figure 24: Envisaged pilot plant PCC unit

In Figure 25 an overview is given of the performance of the solvent developed by HTC. The reported results seem to be promising.



Figure 25: performance of RS-1 compared to MEA

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