
Carbon capture: why we need next generation technologies

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For carbon capture to take off commercially, considerable cost reductions are needed. There are a number of next generation technologies under development that appear potentially game-changing, notably those where capture is inherently incorporated in the power generation process, eg chemical looping and the Allam Cycle. For the existing fleet there is considerable scope for improving the energy efficiency of post-combustion processes by making use of lower grade heat and/or reclaiming energy usually lost elsewhere in the process. But these advanced technologies will need substantial government support if they are to progress beyond the pilot stage. By Toby Lockwood, IEA Clean Coal Centre, UK

As the first full-scale application of carbon capture and storage (CCS) to a power plant, the opening of Canada's Boundary Dam plant in 2014 was heralded by the emerging CCS industry as a landmark moment which would lead to similar developments worldwide. However, other commercial demonstrations of the technology in power generation have so far remained limited to the Petra Nova and Kemper Country projects under construction in the USA – both scheduled to start early next year. Like Boundary Dam, these projects share the favourable circumstances of a cheap and plentiful coal supply and a market for CO₂ for use in enhanced oil recovery. Meanwhile in Europe, where neither of these drivers are usually present, the development of CCS has largely stalled, with the cancellation of the UK CCS demonstration programme late last year putting an end to one of the last major prospects in the region.

The need for cost reduction

Despite the demonstrated technical viability of CCS for coal plant applications, it is clear that the huge economic cost of the technology remains a deterrent to investment in all but the most favourable regulatory and economic environments. As for most low-carbon energy sources, some form of government support in the form of CO₂ pricing or guaranteed power price will be a necessary incentive for CCS, but the cost-of-electricity increase of up to 80% and CO₂ capture price of US\$60/t estimated for state-of-the-art technologies appear beyond the level most governments are willing to support. A considerable research effort worldwide has therefore aimed to develop a second-generation of lower cost CO₂ capture technologies which can make CCS a more competitive option for low-carbon energy. In particular, the US Department of Energy has funded a wide-reaching research programme to investigate new concepts at all levels of technical readiness, but research is also active at a number of academic institutes and private sector companies throughout the developed world, commonly setting cost targets of around US\$30/t CO₂ or a 30% cost-of-electricity reduction on existing technologies.

Three fundamentally different strategies for CO₂ capture from power plants are all considered to be sufficiently developed in some form for full-scale demonstration, with post-combustion capture with amine solvents employed by Boundary Dam and the Petra Nova project in Texas, solvent-based pre-combustion capture selected by the Kemper County plant in Mississippi, and several planned oxyfuel combustion plants falling at the final hurdle due to

insufficient funds, including White Rose in the UK and Futuregen 2.0 in the US. In their current forms, each of these approaches impose a significant energy penalty on the power plant which can account for up to a third of the gross output, requiring a much larger plant to generate the same amount of power and, together with the substantial cost of the capture plant itself, accounting for most of the cost of electricity increase. Researchers have therefore faced the task of developing much more efficient processes without using excessively costly materials or equipment – a particularly challenging prospect in the context of coal combustion, where the presence of corrosive species and huge volumes of gas to process make for demanding operating conditions and require either high throughput processes or very large equipment.

Post-combustion, reducing the energy requirements

Nearly all carbon capture strategies are faced with an inherent gas separation step which is usually the most energy demanding part of the process, but which should also be theoretically achievable at much higher efficiencies than currently reached. In post-combustion capture, where CO₂ is removed from standard flue gases consisting largely of nitrogen, commercial processes use amine-based aqueous solvents which strongly react with CO₂ and consequently need to be heated with steam to regenerate the solvent. New solvent systems have attempted to reduce the large energy requirement of this process either by reducing the strength of the interaction with CO₂ or reducing the thermal mass of the CO₂-rich product to be heated, ideally whilst using more environmentally benign chemicals such as simple metal carbonates or amino acids. Much academic research into the use of ionic liquids to optimise the reaction enthalpy has largely failed to progress due to excessive costs and issues with high viscosity. Research by GE, IFPE, the University of Melbourne and others has looked at biphasic systems, where the reaction with CO₂ produces a separate liquid or solid phase which can be easily separated from the rest of the solvent, resulting in a lower mass to be heated. However, many of these concepts appear to offer only incremental cost reductions and have yet to be tested at a large pilot scale.

Perhaps most promisingly, engineered forms of the enzyme carbonic anhydrase can be used to accelerate the usually slow reaction of CO₂ with low cost carbonate solvents. Using a free-flowing enzyme that permits the use of low grade heat in solvent stripping, Canadian company CO₂ Solutions has operated a 10 tpd pilot and projects capture costs below 40 US\$/t CO₂.

Solid sorbents are also widely used in other gas separation processes such as air drying and separation, and have been extensively investigated for post-combustion capture from coal flue gas. Whilst higher CO₂ capacities and lower regeneration energies than solvent systems are theoretically possible with CO₂-selective sorbents, managing heat transfer in large-scale solid systems is challenging. A few large pilot plants have been successfully operated using pressure swing absorption cycles with simple sorbents such as activated carbons and zeolites, but most current research has instead focussed on temperature swing processes which avoid energetically demanding vacuum pumping and are able to use more selective sorbents that chemically react with the CO₂. Fluidised bed reactors with sorbent pellets are an effective means of achieving the necessary rates of heat transfer in temperature swing processes, with Korean utility KEPCO having operated a 10 MW equivalent facility since 2013 using a simple sodium carbonate material. Fixed bed reactors avoid the sorbent attrition inherent in fluidised beds and offer more straightforward operation, but structured materials and monoliths are required for sufficiently rapid heat and mass transfer in the bed.

Based on the rotary heat exchangers familiar to the power industry, a novel concept from Inventys uses a structured carbon sorbent which continuously rotates between flows of flue gas, regenerating steam, and cooling air. By retaining some of the heat released by CO₂ adsorption, this efficient process is projected to be capable of achieving below US\$30/t and is in line to receive US DOE funding towards a 10 MW equivalent pilot plant (Figure 1).

CO₂-selective polymer membranes offer an alternative post-combustion capture solution that avoids steam extraction or chemical waste, and could be scaled up in a straight-forward modular fashion, although without economies of scale. Despite a great deal of materials research focussing on developing membranes sufficiently selective to produce high purity CO₂ in a single separation step, a more fundamental limitation on separation performance is imposed by the limited pressure gradients that can practically be used to drive the process. To help relax this constraint, US manufacturer Membrane Technology and Research has proposed a two-stage system where combustion air is used as a sweep gas to drive the second stage and increase the CO₂ concentration at the first stage (Figure 2). Recycling CO₂ to the boiler in this way lowers the flame temperature without diminishing the efficiency of power generation, as high boiler temperatures are never fully exploited by the steam cycle. Capture costs achievable with such a process are still limited to >US\$40/t, but become much more competitive if CO₂ capture rates are relaxed to below 90%. Lowering the cost of membrane processes further will rely on the development of highly permeable materials which retain sufficient selectivity, for which the use of inorganic-organic composite materials shows some potential.

Some notable post-combustion capture concepts allow the capture plant to generate its own power, significantly mitigating the energy consumption of the gas separation step. Developed to a scale of 1-2 MWth by separate EU research consortiums known as CaOling and Scarlet, calcium looping is a sorbent-based process featuring this advantage, based on the carbonation reaction of CaO. The resulting limestone requires direct heating in its own oxyfuel-fired boiler to regenerate the sorbent and release CO₂, generating additional power and regaining the energy lost in carbonation.

Gas-fuelled molten carbonate fuel cells also separate CO₂ from flue gases as part of their power generation process, and are currently being scaled up to 10 MW equivalent by US company FuelCell Energy.

Pre-combustion, realising its full potential

Pre-combustion capture is associated with integrated gasification combined cycle power plants, in which coal is first gasified to CO and hydrogen prior to combustion in a gas turbine. Whilst the handful of early demonstration plants have now mostly closed, the technology is being revived in the US, China, and Japan with a view to realising its potential for higher efficiency carbon capture than post-combustion processes.

Once the water gas shift reaction has converted CO to CO₂ and more hydrogen, the syngas contains a much higher partial pressure of CO₂ than coal flue gas, allowing for easier separation and more effective use of pressure-driven systems such as sorbents and membranes. On the other hand, the conventional process is complex and comes with several energy losses besides the relatively efficient solvent-based capture step. Most research aims to avoid unnecessary cooling of the syngas with the use of sorbents or membranes which can operate at warm syngas temperatures, so that the heat and water content can be retained in the hydrogen-rich gas fed to the turbine. In particular, the CO₂ capture step could be integrated into the same reactor as the water gas shift, helping drive the equilibrium reaction to completion and

reducing the need for steam reagent. The EU project CAESAR developed such a sorbent-enhanced water gas shift process using a pressure swing cycle and a hydrotalcite sorbent, currently being scaled up as part of a follow up project targeted at blast furnaces, and targeting a capture cost of US\$25/t. Membrane reactors can also be employed in this fashion, with considerable research on the application of hydrogen selective materials which are viable at high temperatures such as palladium alloys and carbon molecular sieves, but these tests remain at relatively small scale.

Oxyfuel options

The most recent of the three capture strategies, the principle of oxyfuel combustion is to fire coal in oxygen rather than air, producing a stream of mostly CO₂ and water from which CO₂ can be purified relatively easily. Whilst conventional oxyfuel usually recycles flue gas to the boiler to mimic the temperatures and heat transfer rates typical of air combustion, advanced oxyfuel research has largely aimed to exploit the properties of oxyfuel combustion in novel, higher efficiency power generation systems which can offset the considerable energy penalty of oxygen production from air. The most widely investigated approach is to carry out combustion at high pressures, enabling the significant latent heat of water vaporisation in the flue gases to be recovered as useful work, along with other benefits including smaller equipment, reduced air ingress, and improved heat transfer. Early development by Italian companies ENEL and ITEA led to a small pilot being commissioned in 2007, whilst work at Washington University has conceived a more advanced concept with minimal flue gas recycle based on several connected furnace stages to which fuel is added incrementally. A scaled up version of this process is calculated to have an efficiency penalty below 4% points, but only a small combustor pilot has so far been trialled.

As a potentially effective alternative to pre-combustion capture, oxyfuel combustion can also be applied to gas turbines running on coal syngas, with a number of different cycles having been developed and tested on natural gas. The Allam Cycle process from 8 Rivers Capital is particularly noteworthy, as even with coal gasification incorporated it is estimated to be capable of higher efficiencies (>50%, LHV) and lower electricity costs than a conventional coal plant without CO₂ capture (Figure 3). Syngas and oxygen are combusted at 300 bar to produce an exhaust of supercritical CO₂ and steam which can be used to drive a highly efficient and compact turbine. Under a separate entity known as NET Power, a natural gas-based version of the process is currently being scaled up to a 50 MWt pilot in Texas and expected to begin commissioning early next year. For its application to coal syngas, the company is working on a modified combustor and an integrated SO_x/NO_x removal process based on the lead chamber reactions.

Chemical looping combustion is a unique approach to carbon capture which avoids any gas separation process and is therefore theoretically capable of achieving very low energy penalties which are mainly associated with CO₂ compression. Often regarded as a variant of oxyfuel combustion, oxygen is delivered to the fuel by a solid oxide known as the carrier material, using interconnected reactors to circulate the material between the fuel and an air reactor which regenerates the oxide. As solid-solid reactions are much too slow, chemical looping combustion of coal requires in-situ gasification with fluidising steam or CO₂, and the inevitable loss of oxygen carrier with ash removal means that low cost carriers are also a necessity. Alstom (now GE) has commissioned the largest chemical looping combustion pilot, 3 MWt, using interconnected fluidised beds and a carrier based on calcium sulphate, which is generated in situ from limestone.

Meanwhile, Babcock and Wilcox has developed a relatively compact looping process based on countercurrent moving bed reactors and using iron oxide as a low cost carrier material, which has so far been tested at a relatively small pilot scale.

Both these technologies estimate net efficiencies of over 35% (HHV) with capture costs below US\$30/t, and have recently received US DOE funding to accelerate their scale-up to 10 MW.

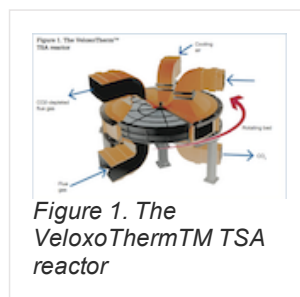
Promising technologies need support

Technologies which inherently incorporate CO₂ capture into the power generation process, such as chemical looping combustion and the Allam Cycle, appear the most promising options for any future coal plants. Both these technologies appear capable of achieving a step change reduction in capture costs, which could greatly accelerate uptake, and are currently undergoing active scale up to large pilot plants.

For the world's vast existing coal fleet, some form of post-combustion capture will still be necessary, and any prospective technologies may need to demonstrate significant gains in order to disrupt the growing market dominance of amine solvents. Nevertheless, through fundamental innovations such as the use of lower grade heat or reclaiming energy usually lost elsewhere in the process, a number of concepts appear to offer much higher efficiencies and corresponding cost reductions to US\$30/t or less.

Considerable variation in site-specific factors such as environmental regulations, cost of capital and labour, or water availability, mean that no single technology will always be optimum, and a range of solutions should be developed. A key example is the growing trend for legislation to require only partial capture on coal plant, which may begin to place membrane-based technologies in a much more favourable light than the conventional target of 90% capture.

Whilst development of these second and third generation technologies may prove to be necessary for CCS deployment to pick up in the power sector, their implementation will be much more straightforward if greater experience is first gained with existing technologies and an early CO₂ transport and storage infrastructure is developed. With even the most advanced of these concepts likely to be several years from full-scale demonstration, there is a risk of the industry stagnating and research funding waning in the absence of more visible CCS projects. As ever, considerable government support will be required if even the most promising of these technologies are to overcome the well-known economic barriers to commercial deployment, and many will not progress beyond the pilot scale.



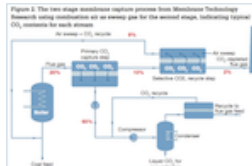


Figure 2. The two stage membrane capture process from Membrane Technology Research using combustion air as sweep gas for the second stage, indicating typical CO₂ contents for each stream

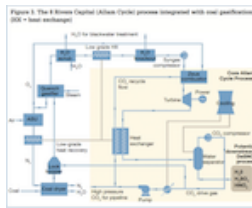


Figure 3. The 8 Rivers Capital (Allam Cycle) process integrated with coal gasification (HX = heat exchange)



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