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Review

The outlook for improved carbon capture technology

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ABSTRACT

Carbon capture and storage (CCS) is widely seen as a critical technology for reducing atmospheric emissions of carbon dioxide (CO_2) from power plants and other large industrial facilities, which are major sources of greenhouse gas emissions linked to global climate change. However, the high cost and energy requirements of current CO_2 capture processes are major barriers to their use. This paper assesses the outlook for improved, lower-cost technologies for each of the three major approaches to CO_2 capture, namely, post-combustion, pre-combustion and oxy-combustion capture. The advantages and limitations of each of method are discussed, along with the current status of projects and processes at various stages in the development cycle. We then review a variety of "roadmaps" developed by governmental and private-sector organizations to project the commercial roll-out and deployment of advanced capture technologies. For perspective, we also review recent experience with R&D programs to develop lower-cost technologies for SO_2 and NO_x capture at coal-fired power plants. For perspective on projected cost reductions for CO_2 capture we further review past experience in cost trends for SO_2 and NO_x capture systems. The key insight for improved carbon capture technology is that achieving significant cost reductions will require not only a vigorous and sustained level of research and development (R&D), but also a substantial level of commercial deployment, which, in turn, requires a significant market for CO_2 capture technologies. At present such a market does not yet exist. While various incentive programs can accelerate the development and deployment of improved CO_2 capture systems, government actions that significantly limit CO_2 emissions to the atmosphere ultimately are needed to realize substantial and sustained reductions in the future cost of CO_2 capture.

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1. Introduction

Global climate change is an issue of major international concern and the focus of proposed mitigation policy measures in the U.S. and elsewhere. In this context, the technology of carbon capture and storage (CCS) has received increasing attention over the past decade as a potential method of limiting atmospheric emissions of carbon dioxide (CO_2)—the principal “greenhouse gas” linked to climate change.

Worldwide interest in CCS stems principally from three factors. First is growing recognition that large reductions in global CO_2 emissions are needed to avoid serious climate change impacts [1,2]. Because electric power plants are a major source of CO_2 emissions, those emissions must be curtailed significantly.

Second is the realization that large emission reductions cannot be achieved easily or quickly simply by using less energy or by replacing fossil fuels with alternative energy sources that emit little or no CO_2. The reality is that the world today relies on fossil fuels for

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over 85% of its energy use (as does the U.S.). Changing that picture dramatically will take time. CCS thus offers a way to get large CO_2 reductions from power plants and other industrial sources until cleaner, sustainable energy sources and technologies can be widely deployed.

Finally, energy-economic models show that adding CCS to the suite of other GHG reduction measures significantly lowers the cost of mitigating climate change. Such studies also indicate that by 2030 and beyond CCS is a major component of a cost-effective portfolio of emission reduction strategies [3,4].

Fig. 1 depicts the overall CCS process applied to a power plant or other industrial process. The CO_2 produced from carbon in the fossil fuels or biomass feedstock is first captured, and then compressed to a dense (supercritical) fluid to facilitate its transport and storage. The main storage option is underground injection into a suitable geological formation. The dominant transport mode for supercritical CO_2 is a pipeline.

At the present time, CCS is not yet commercially demonstrated in the primary application for which it is envisioned—large-scale electric power plants fueled by coal or natural gas. Furthermore, the cost of CCS today is relatively high, due mainly to the high cost of CO_2 capture (which includes the cost of CO_2 compression needed for transport and storage). This has prompted a variety of governmental and private-sector research programs in the U.S. and elsewhere focused on developing more cost-effective methods of CO_2 capture.

1.1. Objectives and scope of this paper

The objective of this paper is to provide a realistic assessment of the outlook for improved, lower-cost CO_2 capture systems for use at power plants and other industrial processes. Issues and technologies associated with CO_2 transport and storage are thus outside the scope of this paper. More specifically we seek to:

- Describe each of the three current approaches to CO_2 capture, namely, (a) post-combustion capture of flue gas CO_2 via chemical treatment; (b) pre-combustion chemical removal of CO_2 from the synthesis gas produced in an integrated coal gasification combined cycle (IGCC) power plant; and (c) oxy-combustion systems that produce a flue gas with high CO_2 concentrations amenable to capture without a post-combustion chemical process.
- Review and summarize current research on carbon capture in the United States and elsewhere to assess the likely evolution of CO_2 capture technologies and the potential for new "breakthrough technologies" such as novel solvents, sorbents, membranes and thin films for gas separation. This discussion also will identify where various technologies lie in the research and development (R&D) "pipeline" in order to better assess their prospects and timetable for commercial availability.
- Review the roll-out schedules for advanced capture technologies currently anticipated by government agencies, as well as recent historical trends in the development, commercialization and cost of other types of new power plant capture technologies. This analysis is intended to provide a historical perspective to help judge the pace at which significant cost reductions and gains in CO_2 capture efficiency can be reasonably expected and the key drivers that influence the pace of innovation.

1.2. Organization of this paper

Consistent with the above objectives, Section 2 first gives an overview of CO_2 capture technologies, their application to new and existing facilities, and their current costs. Section 3 then discusses the process of technological change and defines five stages of technological development used in this paper to describe the status of CO_2 capture technologies. Section 4 elaborates on each of the three major categories of CO_2 capture systems (post-combustion, pre-combustion and oxy-combustion capture). For each category, the current status of technologies in the R&D pipeline is described along with the technical challenges that must be overcome to move forward. Sections 5 and 6 then discuss the prospects for improved, lower-cost capture technologies and the timetables for commercialization projected by governmental and private-sector organizations involved in capture technology R&D. For perspective, Section 7 looks retrospectively at recent experience for the pace of technology innovation and deployment for other power plant emission control systems. It also discusses some of the key drivers of technology innovation that influence the future prospects for carbon capture systems. Finally, Section 8 summarizes key findings and conclusions.

2. Overview of CO_2 capture technologies

A variety of technologies for separating (capturing) CO_2 from a mixture of gases are commercially available and widely used

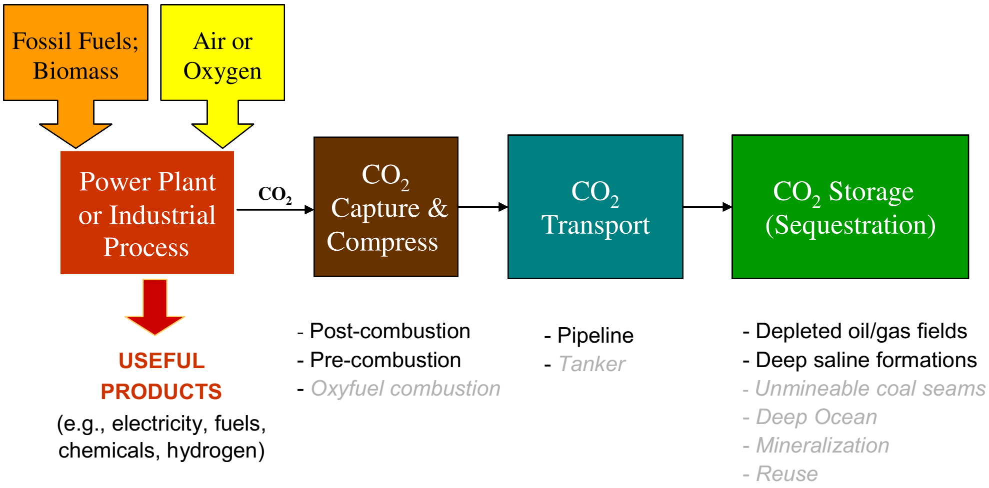

Fig. 1. Schematic of a CCS system, consisting of CO_2 capture, transport and storage. Carbon inputs may include fossil fuels and biomass. Technical options are listed below each stage. Those in italics are not yet available or deployed at a commercial scale [5].

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today, typically as a purification step in an industrial process. Fig. 2 illustrates the variety of technical approaches available. The choice of technology is dependent on the requirements for product purity and on the conditions of the gas stream being treated (such as its temperature, pressure CO_2, concentration and the type and level of trace species or impurities). Common applications for CO_2 capture systems include the removal of CO_2 impurities in natural gas treatment and the production of hydrogen, ammonia and other industrial chemicals. In most cases, the captured CO_2 stream is simply vented to the atmosphere. In a few cases it is used in the manufacture of other chemicals [7].

CO_2 also has been captured from a portion of the flue gases produced at power plants burning coal or natural gas. Here, the captured CO_2 is sold as a commodity to nearby industries such as food processing plants. Globally, however, only a small amount of CO_2 is utilized to manufacture industrial products and nearly all of it is soon emitted to the atmosphere (for example, from carbonated drinks).

Since most anthropogenic CO_2 is a byproduct of the combustion of fossil fuels, CO_2 capture technologies, in the context of CCS, are commonly classified as either pre-combustion or post-combustion systems, depending on whether carbon (in the form of CO_2) is removed before or after a fuel is burned. A third approach, called oxyfuel or oxy-combustion, does not require a CO_2 capture device. This concept is still under development and is not yet commercial in power plant operations, although some industrial processes do employ oxygen combustion (e.g., in the glass and metals industries), albeit without separating CO_2 from the gas stream. Industrial processes that do not involve combustion employ the same types of CO_2 capture systems that would be employed at power plants.

In all cases, the aim is to produce a stream of pure CO_2 that can be permanently stored or sequestered, typically in a geological formation. This requires high pressures to inject CO_2 deep underground. Thus, captured CO_2 is first compressed to a supercritical state, where it behaves as a liquid that can be readily transported via pipeline and injected into a suitable geological formation. The CO_2 compression step is commonly included as part of the capture system since it is usually located at the industrial plant site where CO_2 is captured.

2.1. Post-combustion processes

As the name implies, these systems capture CO_2 from the flue gases produced after fossil fuels or other carbonaceous materials (such as biomass) are burned. Combustion-based power plants provide most of the world's electricity today. In a modern coal-fired power plant, pulverized coal (PC) is mixed with air and burned in a furnace or boiler. The heat released by combustion generates steam, which drives a turbine-generator (Fig. 3). The hot combustion gases exiting the boiler consist mainly of nitrogen (from air) plus smaller concentrations of water vapor and CO_2 formed from the hydrogen and carbon in the fuel. Additional products formed during combustion from impurities in coal include sulfur dioxide, nitrogen oxides and particulate matter (fly ash). These regulated air pollutants, as well as other trace species such as mercury, must be removed to meet applicable emission standards. In some cases, additional removal of pollutants (especially SO_2) is required to provide a sufficiently clean gas stream for subsequent CO_2 capture.

With current technology, the most effective method of CO_2 capture from the flue gas of a PC plant is by chemical reaction with an organic solvent such as monoethanolamine (MEA), one of a family of amine compounds. In a vessel called an absorber, the flue gas is "scrubbed" with an amine solution, typically capturing 85 to 90 percent of the CO_2. The CO_2-laden solvent is then pumped to a second vessel, called a regenerator or stripper, where heat is applied in the form of steam to release the CO_2. The resulting stream of concentrated CO_2 is then compressed and piped to a storage site, while the depleted solvent is recycled back to the absorber. Fig. 4 shows details of a typical post-combustion capture system design.

The same post-combustion capture technology that would be used at a PC plant also would be used for post-combustion CO_2 capture at a natural gas-fired boiler or combined cycle (NGCC) power plant, as depicted in Fig. 5. Although the flue gas CO_2 concentration is more dilute than in coal plants, high removal efficiencies can still be achieved with amine-based capture systems. The absence of impurities in natural gas also results in a clean flue gas stream, so that no additional cleanup is needed for effective CO_2 capture. Further details on the design, performance and operation of amine-based capture technologies can be found in the technical literature [6,7,9].

2.2. Pre-combustion CO_2 capture processes

To remove carbon from fuel prior to combustion it must first be converted to a form amenable to capture. For a coal-fueled plant,

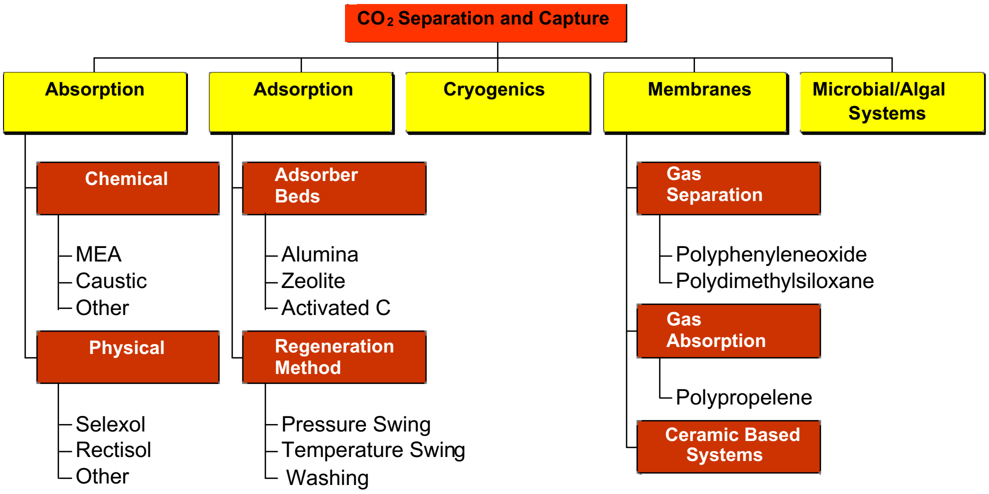

Fig. 2. Technical options for CO_2 capture. The choice of method depends strongly on the particular application [6].

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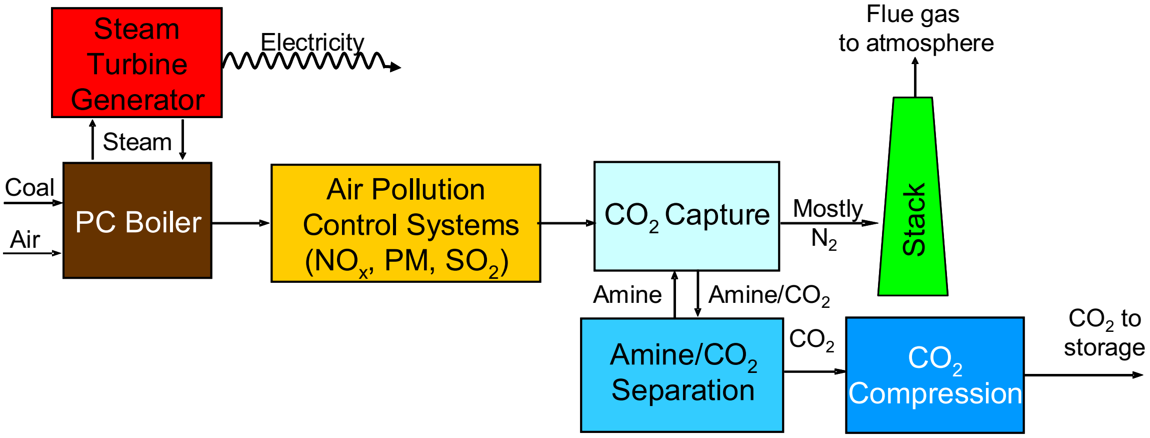

Fig. 3. Simplified schematic of a coal-fired power plant with post-combustion CO_2 capture using an amine scrubber system. Other major air pollutants (nitrogen oxides, particulate matter and sulfur dioxide) are removed from the flue gas prior to CO_2 capture [8].

this is accomplished by reacting coal with steam and oxygen at high temperature and pressure, a process called partial oxidation, or gasification. The result is a gaseous fuel consisting mainly of carbon monoxide and hydrogen—a mixture known as synthesis gas, or syngas—which can be burned to generate electricity in a combined cycle power plant similar to the NGCC plant described above. This approach is known as integrated gasification combined cycle (IGCC) power generation. After particulate impurities are removed from the syngas, a two stage shift reactor converts the carbon monoxide to CO_2 via a reaction with steam (H_2O). The result is a mixture of CO_2 and hydrogen. A chemical solvent, such as the widely used commercial product Selexol (which employs a glycol-based solvent), then captures the CO_2, leaving a stream of nearly-pure hydrogen that is burned in a combined cycle power plant to generate electricity, as depicted in Fig. 6.

Although the fuel conversion steps of an IGCC plant are more elaborate and costly than traditional coal combustion plants, CO_2 separation is much easier and cheaper because of the high operating pressure and high CO_2 concentration of this design. Thus, rather than requiring a chemical reaction to capture CO_2 (as with amine systems in post-combustion capture), the mechanism employed in pre-combustion capture involves physical absorption into the solvent, followed by release of the CO_2 when the sorbent pressure is dropped, typically in several stages, as depicted in Fig. 7. Nonetheless, there is still a significant energy penalty associated with CO_2 capture due to the need for a shift reactor and other processes, as elaborated below.

Pre-combustion capture also can be applied to power plants using natural gas. As with coal, the raw gaseous fuel is first converted to syngas via reactions with oxygen and steam—a process called reforming. This is again followed by a shift reactor and CO_2 separation, yielding streams of concentrated CO_2 (suitable for storage) and hydrogen. This is the dominant method used today to manufacture hydrogen. If the hydrogen is burned to generate electricity, as in an IGCC plant, we have pre-combustion capture. While this is usually more costly than post-combustion capture for natural gas-fired plants, some power plants of this type have been proposed [11]. Details regarding the design, performance and

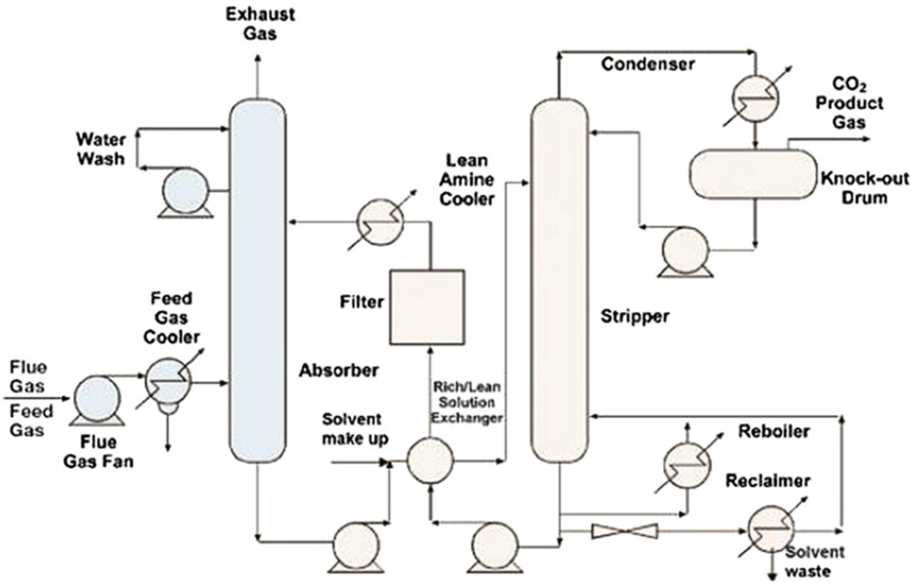

Fig. 4. Details of flue gas and sorbent flows for an amine-based post-combustion CO_2 capture system, showing the absorber (on the left) and regenerator (on the right) [7].

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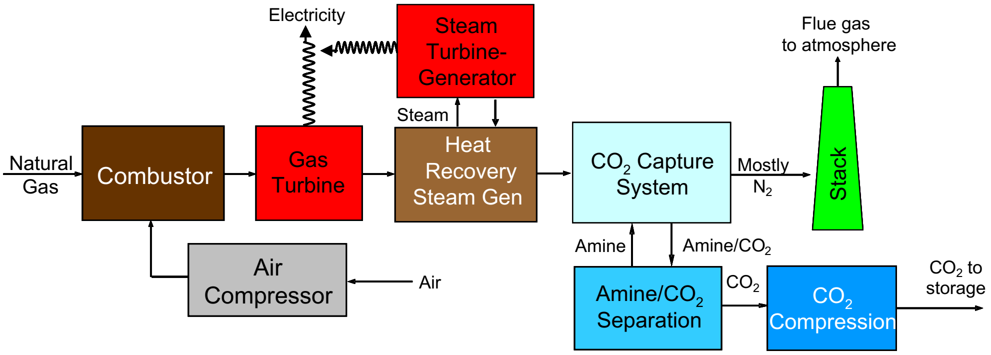

Fig. 5. Simplified schematic of an amine-based post-combustion CO_2 capture system applied to a natural gas combined cycle (NGCC) power plant [8].

operation of pre-combustion capture systems can be found in other studies [7,12].

2.3. Oxy-combustion systems

Oxy-combustion (or oxyfuel) systems are being developed as an alternative to post-combustion CO_2 capture for conventional coal-fired power plants. Here, pure oxygen rather than air is used for combustion. This eliminates the large amount of nitrogen in the flue gas stream. After the particulate matter (fly ash) is removed, the flue gas consists only of water vapor and CO_2 plus smaller amounts of pollutants such as sulfur dioxide (SO_2) and nitrogen oxides (NO_x). The water vapor is easily removed by cooling and compressing the flue gas. Additional removal of air pollutants leaves a nearly-pure CO_2 stream that can be sent directly to storage, as depicted in Fig. 8.

The principal attraction of oxy-combustion is that it avoids the need for a costly post-combustion CO_2 capture system. Instead, however, it requires an air separation unit (ASU) to generate the relatively pure (95–99 percent) oxygen needed for combustion. Roughly three times more oxygen is needed for oxyfuel systems than for an IGCC plant of comparable size, so the ASU adds significantly to the cost. Typically, additional flue gas processing also is needed to reduce the concentration of conventional air pollutants, so as to comply with applicable environmental standards; to prevent the undesirable buildup of a substance in the flue gas recycle loop; or to achieve pipeline CO_2 purity specifications (whichever requirement is the most stringent). Because combustion temperatures with pure oxygen are much higher than with air, oxy-combustion also requires a large portion (roughly 70 percent) of the inert flue gas stream to be recycled back to the boiler in order to maintain normal operating temperatures, although for new oxy-fueled boilers designs have been proposed to reduce or eliminate external recycle through means such as slagging combustors or controlled staging of non-stoichiometric burners. To avoid unacceptable levels of oxygen and nitrogen in the flue gas, the system also has to be carefully sealed to prevent any leakage of air into the flue gas. This is a challenge since such leakage commonly occurs at flanges and joints along the flue gas ducts of existing power plants, especially as plants age.

As a CO_2 capture method, oxy-combustion has been studied in laboratory and pilot plant facilities at scales of 30 $MW_{thermal}$ (equivalent to about 10 MW_{elec}). A variety of designs have been proposed for commercial scale systems and large-scale demonstration projects are now planned [7]. Although oxyfuel systems can theoretically capture all of the CO_2 produced, the need for additional gas treatment systems to remove impurities decreases the capture efficiency to about 90 percent in most current designs.

In principle, oxy-combustion also can be applied to simple cycle and combined cycle power plants fueled by natural gas or distillate oil. These conceptual designs are discussed more fully in Section 4.3. As a practical matter, however, they would require significant and costly modifications to the design of current gas turbines and other plant equipment, with uncertain (and likely limited) market potential for greenhouse gas abatement. Thus, the current focus of oxy-combustion development is on coal-fired power plant applications.

2.4. Capture system energy penalty

The energy requirements of current CO_2 capture systems are roughly ten to a hundred times greater than those of other environmental control systems employed at a modern electric power

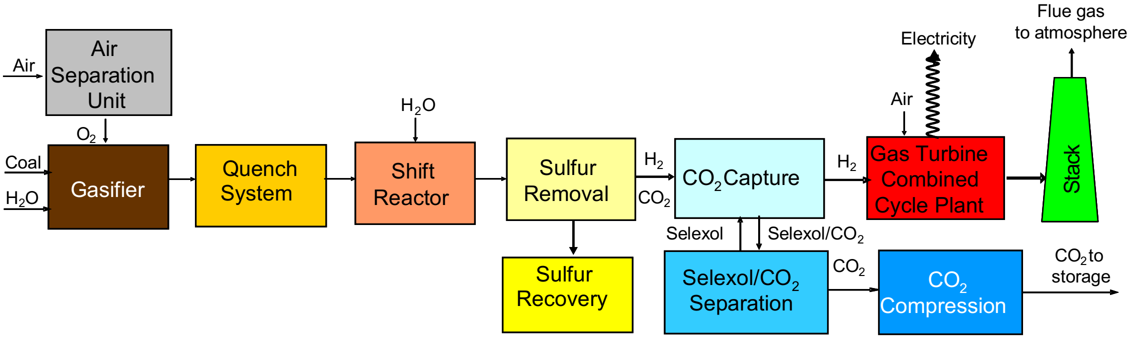

Fig. 6. Simplified schematic of an integrated gasification combined cycle (IGCC) coal power plant with pre-combustion CO_2 capture using a water-gas shift reactor and a Selexol CO_2 separation system [8].

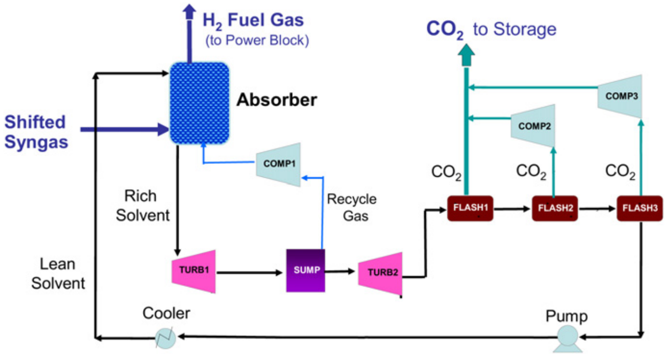

Fig. 7. Details of flue gas and sorbent flows for pre-combustion CO_2 capture [10].

plant. This energy “penalty” lowers the overall (net) plant efficiency and significantly increases the net cost of CO_2 capture. Table 1 shows that of the three CO_2 capture approaches discussed earlier, post-combustion capture on a subcritical (low-efficiency) PC plant is the most energy-intensive, requiring more than twice the energy per unit of electricity output as pre-combustion capture on a new IGCC plant. The table also shows the corresponding reduction in net plant output for a fixed energy input to a plant—a definition of energy penalty also used in the literature and best suited for retrofit situations where plant energy input is fixed.

Lower plant efficiency means that more fuel is needed to generate electricity relative to a similar plant without CO_2 capture. For coal combustion plants, this means that proportionally more solid waste is produced and more chemicals, such as ammonia and limestone, are needed (per unit of electrical output) to control NO_x and SO_2 emissions. Plant water use also increases significantly because of the additional cooling water needed for current amine capture systems. Because of the efficiency loss, a capture system that removes 90 percent of the CO_2 from the plant flue gas winds up reducing the net (avoided) emissions per kilowatt-hour (kWh) by a smaller amount, typically 85 to 88 percent [7].

In general, the higher the power plant efficiency, the smaller are the energy penalty and associated impacts. For this reason, replacing or repowering an old, inefficient plant with a new, more efficient unit with CO_2 capture can still yield a net efficiency gain that decreases all plant emissions and resource consumption. Thus, the net impact of the CO_2 capture energy penalty must be assessed in the context of a particular situation or strategy for reducing CO_2 emissions. Innovations that raise the efficiency of power generation also can reduce the impacts and cost of carbon capture. Table 2 shows that the overall energy requirements for PC and IGCC plants is divided between electricity needed to operate fans, pumps and CO_2 compressors, plus thermal energy requirements (or losses) for solvent regeneration (PC plants) and the water-gas shift reaction (IGCC plants). Thermal energy requirements are clearly the largest source of net power losses and the priority area for research to reduce those losses. For oxy-combustion systems, the electrical energy required for oxygen production is the biggest contributor to the energy penalty.

Table 1
Representative values of power plant efficiency and CCS energy penalty. All efficiency values are based on the higher heating value (HHV) of fuel. For each plant type, there is a range of reported efficiencies (and associated energy penalties) around the values shown here [7,13–15].

Power plant and capture system type	Net plant efficiency (%) w/o CCS	Net plant efficiency (%) with CCS	CCS energy penalty	
			Additional energy input (%) per net kWh output[a]	Reduction in net kWh output (%) for a fixed energy input
Existing subcritical PC, post-combustion capture	33	23	43%	30%
New supercritical PC, post-combustion capture	40	31	29%	23%
New supercritical PC, oxy-combustion capture	40	32	25%	20%
New IGCC (bituminous), pre-combustion capture	40	33	21%	18%
New natural gas comb. cycle, post-combustion capture	50	43	16%	14%

[a] This is the definition of energy penalty recommended by the IPCC [7]. It reflects the incremental primary energy needed to supply a unit of electric power (e.g., 1 kW-hour) to the grid.

2.5. Current cost of CO_2 capture

To gauge the potential benefits of advances in carbon capture technology, it is useful to first benchmark the cost of current systems. Here we review recent cost estimates for power plants and other industrial processes employing capture technologies that are commercially deployed or offered.

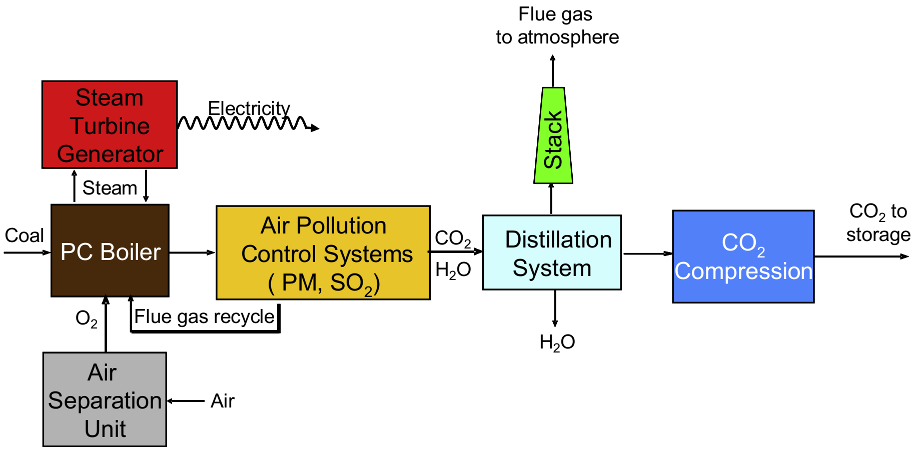

Fig. 8. Simplified schematic of a coal-fired power plant using oxy-combustion technology [8]. Details of plant designs vary across studies. The step shown as a distillation system may include the removal of trace pollutants. Removal of water vapor often is integrated with CO_2 compression.

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Table 2
Break down of the energy penalty for CO_2 capture at supercritical PC and IGCC power plants [13,14].

Energy type and function	Approximate % of total energy penalty
Thermal energy for amine solvent regeneration (post-combustion) or loss in water-gas shift reaction (pre-combustion); or, electricity for oxygen production (oxy-combustion)	~60%
Electricity for CO_2 compression	~30%
Electricity for pumps, fans, etc.	~10%

2.5.1. *Costs for new power plants*

Fig. 9 displays the cost of generating electricity from new power plants with and without CCS, as reported in recent studies based on current commercial post-combustion and pre-combustion capture processes. All plants capture and sequester 90 percent of the CO_2 in deep geologic formations.

The total cost of electricity generation (COE, in $/MWh) is shown as a function of the CO_2 emission rate (tonnes CO_2/MWh) for power plants burning bituminous coal or natural gas. The COE includes the costs of CO_2 transport and storage, but most of the cost (80–90 percent) is for capture (including compression).

The dominant factors responsible for the broad range of costs for each plant type in Fig. 9 are assumptions about the design, operation and financing of the power plant to which the capture technology is applied. For example, higher plant efficiency, larger plant size, higher fuel quality, lower fuel cost, higher annual hours of operation, longer operating life and lower-cost of capital all reduce both the cost of electricity and the unit cost of CO_2 capture. Assumptions about the CO_2 capture system design and operation further contribute to variations in the overall cost. Assumptions vary across studies, and since no single set of assumptions applies to all situations or all parts of the world there is no universal estimate for the cost of CO_2 capture. Cost ranges would be even broader if additional factors such as a range of coal types or a larger range of boiler efficiencies were considered.

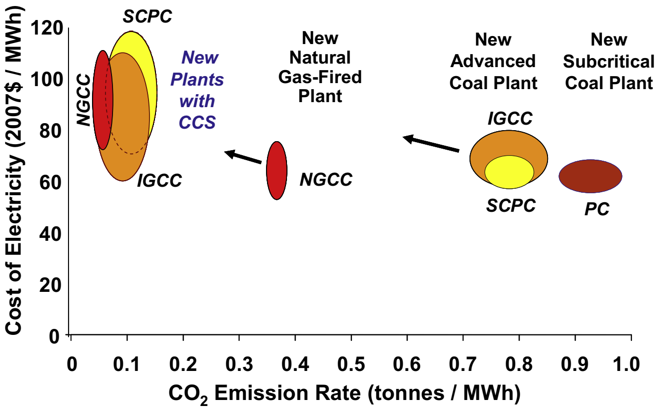

Fig. 9. Cost of electricity generation (constant 2007 US$/MWh) as a function of the CO_2 emission rate (tonnes CO_2/MWh) for new power plants burning bituminous coal or natural gas. PC = subcritical pulverized coal units; SCPC = supercritical pulverized coal; IGCC = integrated gasification combined cycle; NGCC = natural gas combined cycle). Ranges reflect differences in key technical, financial, operational and economic assumptions affecting plant cost, based on data from [5,7,9,13,14,16,17]. Thus, overlapping ovals do not imply that one technology can be more (or less) costly than another under the same set of assumptions (for example, while different studies report overlapping cost values for different SCPC and IGCC plants without CCS, individual studies show IGCC plants to be systematically more costly than a similarly-sized SCPC plant when all other assumptions are held constant).

On a relative basis, CCS is estimated to increase the cost of generating electricity by approximately 60–80 percent at new coal combustion plants and by about 30–50 percent at new coal gasification plants. On an absolute basis, the increased cost translates to roughly $40–70/MWh for supercritical (SCPC) coal plants and $30–50/MWh for IGCC plants using bituminous coal. As noted earlier, the CO_2 capture step accounts for most of this cost.

Fig. 9 also can be used to calculate the cost per tonne of CO_2 avoided for a plant with capture relative to one without. This cost is equivalent to the “carbon price” or CO_2 emissions tax above which the CCS plant is more economical than the plant without capture. For new supercritical coal plants this is currently about $60–80/tonne CO_2. For IGCC plants with and without CCS, the avoidance cost is smaller, about $30–50/tonne CO_2. Since the cost of CO_2 avoided depends on the choice of “reference plant” with no CCS, it is also useful to compare an IGCC plant with CCS to an SCPC reference plant, since without capture SCPC is less expensive than IGCC for the same design premise. In this case the cost of CO_2 avoided increases to roughly $40–60/tonne CO_2. In all cases, costs are lower if the CO_2 can be sold for enhanced oil recovery (EOR) with subsequent geological storage. For plant using low-rank coals (i.e., subbituminous coal or lignite) the avoidance cost may be slightly higher than values based on Fig. 9 [17,18].

2.5.2. *Retrofit costs for existing power plants*

For existing power plants, the feasibility and cost of retrofitting a CO_2 capture system depend heavily on site-specific factors such as the plant size, age, efficiency, the type and design of existing air pollution control systems and availability of space to accommodate a capture unit [6]. In general, the added cost of electricity generation is higher than for a new supercritical plant. A major contributing factor is the lower thermal efficiency typical of existing (subcritical) power plants, which results in a larger energy penalty and higher capital cost per unit of capacity. Other factors include the added capital costs due to physical constraints and site access difficulties during construction of a retrofit project, plus the likely need for upgrades or installation of additional equipment, such as more efficient SO_2 scrubbers needed to meet the low inlet SO_2 specifications for commercial amine systems. The cost per ton of CO_2 avoided also increases as a result of these higher costs.

Studies also indicate that for many existing power plants the most cost-effective strategy for plants that have suitable access to geological storage is to combine CO_2 capture with a major plant upgrade, commonly called repowering. Here, an existing subcritical unit is replaced either by a high-efficiency (supercritical) boiler and steam turbine, or by a gasification combined cycle system [19,20]. In such cases, the cost of CO_2 capture approaches that of a new plant, with some potential savings from the use of existing plant components and infrastructure, as well as from fewer operating permit requirements relative to a new greenfield site.

2.5.3. *Costs for other industrial processes*

There have been far fewer studies of CO_2 capture costs for industrial processes than for power plants. A 2005 study by the Intergovernmental Panel on Climate Change (IPCC) summarized the literature at that time and reported wide ranges in costs within and across industries and processes [7]. More recently, the International Energy Agency (IEA) and United National Industrial Development Organization (UNIDO) assessed the cost of CO_2 capture and storage for several production processes in five major industrial sectors. Those results also show large ranges, as summarized in Table 3.

In general, the incremental cost of capture is lowest in cases where CO_2 is separated as part of the normal process operations, resulting in a stream of high-purity CO_2 that is typically vented and/

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Table 3
Range of cost estimates for CO_2 capture and storage (CCS) at industrial processes, based on [21]. Figures include the cost of CO_2 transport and storage.

Industrial sector	Production processes	Abatement cost (USD/tCO_2 avoided)
High-purity CO_2 sources	Natural gas processing; hydrogen production; ammonia production; ethylene oxide production; Fischer-Tropsch coal-to-liquids	\$30–70
Biomass conversion	Biosynthetic gas; ethanol production; hydrogen production from biomass; biomass-to-liquids; black liquor processing in pulp and paper production	\$35–80
Refineries	Hydrogen production from natural gas steam methane reforming or gasification residues; fluidized catalytic cracking; process heaters	\$45–120
Cement	Dry-process/suspension preheater rotary kiln with or without precalciner/grate cooler	\$55–150
Iron and steel	Blast furnace (pig iron); direct reduced iron; FINEX and HIsarna steelmaking processes	\$60–80

or used in another chemical process (such as to manufacture urea). Such processes include the production of hydrogen from natural gas as well as the purification of raw natural gas prior to distribution (since CO_2 is a common impurity). In these cases, the cost of CO_2 capture and storage is simply the added costs of compression, transport and geological storage. For other industrial processes, CO_2 capture costs depend strongly on a host of site-specific factors that influence the technical, economic and financial parameters that underlie any cost analysis. The cost of CO_2 avoided is highly sensitive to such factors as well as to the choice of a reference plant (which is typically a similar facility without CCS). The references cited earlier discuss in more detail the technologies and assumptions underlying the range of costs reported in Table 3.

2.5.4. Important caveat concerning costs

Construction costs for power plants and industrial equipment escalated dramatically from about 2004 to 2008, as did fuel prices, especially natural gas. Most prices then stabilized or receded during the subsequent economic recession in the U.S. and many other countries. Uncertainty about future trends in material and labor costs, together with the absence of full-scale projects, further clouds the “true” cost of facilities with or without CCS. For power plants, the relative costs of PC and IGCC plants also can change with coal type, operating hours, cost of capital and many other factors [17]. Experience with IGCC power plants is still quite limited and neither PC nor IGCC plants with CCS have yet been built and operated at full-scale. Thus, neither the absolute nor relative costs of these systems can be stated with a high degree of confidence at this time.

2.6. Cost of advanced CO_2 capture processes

A number of cost estimates also are available for many of the “advanced” capture processes discussed later in this paper. Such estimates typically report or anticipate lower costs than the technologies currently in use or offered with commercial guarantees. However, as elaborated later in Section 5, cost estimates for processes in the early stages of development, prior to commercialization, have historically been unreliable and typically have under-estimated true commercial costs. For this reason we choose in this paper to focus on descriptions of these advanced technologies and their hoped-for advantages, recognizing that the impetus for their development is the promise of an improved, lower-cost technology relative to what is currently available. We later use representative cost estimates from the U.S. Department of Energy to illustrate the magnitude of cost reductions foreseen from new technologies.

3. Stages of technology development

The stages of technological development or maturity of carbon capture systems span a broad spectrum. At one end of the spectrum are the current commercial systems described in Section 2. At the opposite end are new concepts or processes that exist only on paper, or perhaps as a small-scale device or experiment in a research laboratory. New or “advanced” technologies commonly seek (and often boast of) higher effectiveness and/or lower-cost than current commercial systems—attributes that are highly desired in the marketplace. At the same time, claims about the cost or performance of processes in the early stages of development are inherently uncertain and subject to change as the technology advances toward commercialization.

In this section we discuss a number of ways to characterize the level of technological development of CO_2 capture systems. The aim is to provide a clear understanding of the steps that are needed to bring a promising new technology to commercial reality. To begin, however, we briefly describe the general process of technological change in order to provide context for a closer examination of innovations in carbon capture technologies.

3.1. The process of technological change

Innovations in carbon capture technology and the commercial adoption of such systems is an example of the general process of technological change. While a variety of terms are used to describe that process, four stages that are commonly defined are:

- Invention: Discovery; creation of knowledge; new prototypes
- Innovation: Creation of a new commercial product or process
- Adoption: Deployment and initial use of the new technology
- Diffusion: Increasing adoption and use of the technology

The first stage is driven by R&D, including both basic and applied research. The second stage—innovation—is a term often used colloquially to describe the overall process of technological change. As used here, however, it refers only to the creation of a product or process that is commercially offered; it does not mean the product will be adopted or become widely used. That happens only if the product succeeds in the final two stages—adoption and diffusion, which reflect the commercial success of a technology innovation.

Studies also show that rather than being a simple linear process, the four stages of technological change are highly interactive, as depicted in Fig. 10. Thus, innovation is stimulated not only by support for R&D, but also by the experience of early adopters, plus added knowledge gained as a technology diffuses more widely into the marketplace. The reductions in product cost that are often observed as a technology matures— commonly characterized as a “learning curve”—reflect the combined impacts of sustained R&D plus the benefits derived from “learning by doing” (economies in the manufacture of a product) and “learning by using” (economies in the operating costs of a product).

This report deals only with the first two stages of Fig. 10 in the context of carbon capture systems at different levels of development or maturity. The goal is to characterize the current status of capture technologies and the outlook for future commercial systems. Later, in Section 6, we discuss the influence of the last two stages (adoption and diffusion) on the pace of innovation and the prospects for lower-cost capture technologies.

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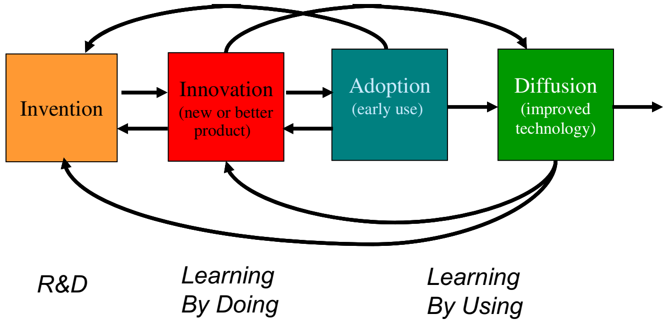

Fig. 10. Stages of technological change and their interactions [22].

3.2. Technology readiness levels (TRLs)

One method of describing the maturity of a technology or system is the scale of Technology Readiness Levels (TRLs) depicted in Fig. 11. First developed for the National Aeronautics and Space Administration (NASA), TRLs were subsequently adopted by the U.S. Department of Defense, as well as by other organizations involved in developing and deploying complex technologies or systems, both in the U.S. and abroad. Recently, researchers at the Electric Power Research Institute (EPRI) also adopted TRLs to describe the status of new post-combustion carbon capture technologies, discussed later in Section 4.1 [23].

The TRL scale has nine levels. At TRL 1 a technology consists only of basic principles, while at TRL 9 it has evolved into a system used successfully in its actual operating environment. TRLs are used to assess the maturity of a technology and the risks of placing it into service for a given mission. A study by the U.S. Government Accountability Office (GAO) found that commercial firms typically do not introduce new technology into a commercial product until it is at the equivalent of TRL 8 or 9, where it has been fully integrated and validated in its working environment. The GAO study also found that a number of government projects it examined tended to be further behind schedule and over budget where unproven technologies were employed, compared to projects designed with more mature technologies [25,26].

The U.S. Department of Energy's Office of Management also recently published a "Technology Readiness Assessment Guide" to provide general guidance as to how critical technologies should be developed before and during their integration into engineered systems [27]. The tailored definitions of TRLs employ four scales of development called lab scale, bench-scale, engineering scale and full-scale (Fig. 12). A technology is considered to be lab scale at TRLs 2 and 3 and bench-scale at TRL 4. The latter is typically a complete system, whereas lab scale involves proof-of-concept for a subsystem or component. A technology at the engineering scale corresponds to TRLs 5 and 6. At TRL 7 and beyond the system is full-scale. Variants of these four categories are used in this report to describe the development stages of carbon capture technologies, as explained below.

3.3. Technology maturity levels used in this study

While the nine level TRL scale is a useful way to describe and compare the status of technologies being considered for deployment in a particular mission or complex system, for purposes of this study, a simpler set of five categories is used to describe the maturity of carbon capture technologies. These five stages reflect not only different levels of maturity but also differences in the physical size and complexity of a CO_2 capture technology at different points in its development. Significant increases in the level of financial commitments also are needed to advance to the final stages of this journey. This representation of "what's in the pipeline" also can convey to policymakers and others the prospects, time requirements and level of financial resources needed to bring improved CO_2 capture systems to the marketplace.

3.3.1. Commercial process

A commercial carbon capture technology or process is one that is available for routine use in a particular application such as a power plant or industrial process. The capture technology is offered for sale by one or more reliable vendors with standard commercial guarantees. As defined here, a commercial technology corresponds to TRL 9, the highest level on the TRL scale. This is the maturity level that electric utility companies normally will require before installing a carbon capture system at a U.S. power plant.

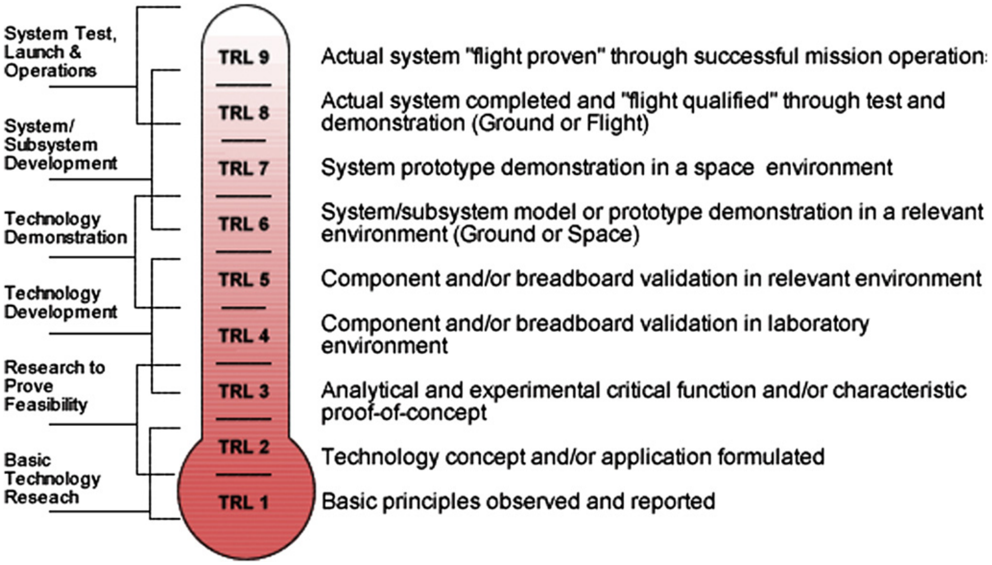

Fig. 11. Descriptions of technology readiness levels (TRLs) [24].

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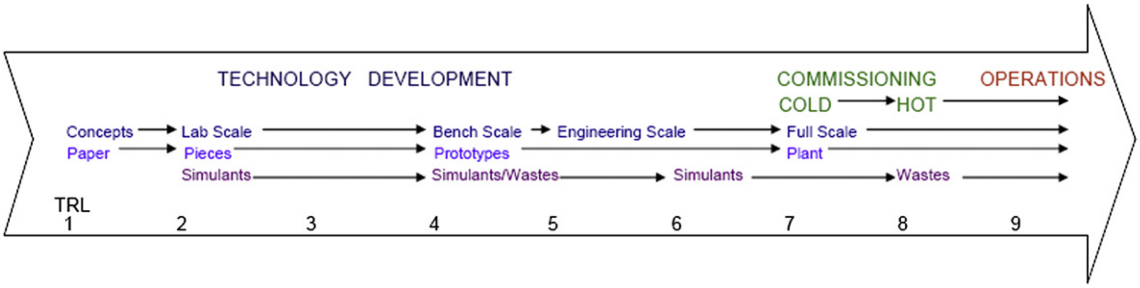

Fig. 12. A U.S. Department of Energy view of technology development stages and their corresponding TRLs [27].

3.3.2. *Full-scale demonstration plant*

The full-scale demonstration stage corresponds to levels 7 and 8 on the TRL scale. It represents the stage at which a CO_2 capture technology is integrated into a full-size system in order to demonstrate its viability and commercial readiness in a particular application. For power plants, such applications might include pulverized coal combustion systems employing oxy-combustion or post-combustion CO_2 capture, as well as IGCC plants employing pre-combustion capture. While there is flexibility in the definition of “full-scale,” in general it would correspond to a gross power plant size of approximately 250 MW or more, with a corresponding CO_2 capture rate of at least 1–2 million tonnes per year for a coal-fired plant. For reference, the median size of U.S. coal-burning power plants today is approximately 650 MW (nameplate capacity). For gas-fired power plants or other industrial applications a full-scale demonstration may have smaller annual quantities of CO_2 captured because of smaller plant sizes and/or lower fuel carbon content.

3.3.3. *Pilot plant scale*

The pilot plant stage is where a process or technology is tested in a realistic environment, but at a scale (and cost) that is typically one to two orders of magnitude smaller than the full-scale demonstration. For carbon capture processes, a pilot plant might be built as a stand-alone facility, or as a unit capturing CO_2 from the slip stream of a full-size power plant. Pilot plants correspond roughly to levels 6 and 7 on the TRL scale. At this stage data are gathered to refine and further develop a process, or to design a full-size (or intermediate size) demonstration plant.

3.3.4. *Laboratory or bench-scale*

The laboratory and bench scales represent the early stage of process development in which an apparatus or process is first successfully constructed and operated in a controlled environment, often using laboratory materials and test gases to simulate a commercial process and flue gas stream. A bench-scale apparatus is typically built as a complete representation of a process or system, whereas laboratory-scale experiments typically seek to validate or obtain data for specific components of a system. Laboratory and bench-scale processes correspond to levels 3, 4 and 5 on the TRL scale.

3.3.5. *Conceptual design*

The conceptual design stage of a CO_2 capture process is one for which the basic science has been developed, but no physical prototypes yet exist. Conceptual designs are often developed and tested with computer models before any laboratory work is done. This allows for confirmation that the design principles are sound, plus some degree of process optimization before progressing to the more expensive laboratory or bench-scale stage. The conceptual design stage corresponds to levels 1 and 2 on the TRL scale.

4. Current status of CO_2 capture technologies

This section of this paper characterizes the current status of carbon capture technologies with respect to the five stages of development outlined above. Each sub-section addresses one of the three main avenues for CO_2 capture, namely, post-, pre- and oxy-combustion systems. Following this we discuss the cost reductions anticipated from advanced capture systems and the projected timetables for their commercialization.

In recent years, carbon capture research and development (R&D) programs have expanded rapidly throughout the world; thus, any summary of “current” activities and projects is soon out of date. For this reason, we make no claims of being comprehensive in our coverage of capture-related R&D activities. Rather, we attempt to synthesize key findings from our own research and from the work of others who track and report on the status of CO_2 capture technology developments. In this regard, we draw heavily upon a set of publicly available databases and CCS project status reports maintained by organizations including the U.S. Department of Energy’s National Energy Technology Laboratory (DOE/NETL), the International Energy Agency’s Greenhouse Gas Control Programme (IEAGHG), the Massachusetts Institute of Technology (MIT) Carbon Sequestration Program and the Global Carbon Capture and Storage Institute (GCCSI) [28–31]. In many cases, the information from public databases has been supplemented by additional data obtained from companies involved in capture technology development and testing.

In each of the sections below our objective is to summarize not only the status of carbon capture technology developments, but also the potential advantages of each new technology, as well as the key technical barriers and challenges that must be overcome to advance the method. Brief descriptions of new processes or capture methods not previously discussed in Section 3 also are provided.

4.1. *Status of post-combustion CO_2 capture*

This section summarizes the status of post-combustion CO_2 capture technologies at various stages of development. While the most advanced systems today employ amine-based solvents, processes at the earliest stages of development employ a variety of novel solvents, solid sorbents and membranes for CO_2 capture or separation.

4.1.1. *Commercial processes*

Although not yet deployed commercially on power plants at full-scale, as noted in Section 2.1, post-combustion CO_2 capture systems have been used commercially for many decades in other industrial processes, mainly for purifying gas streams other than combustion products. The use of amines to capture CO_2 was first patented over eighty years ago. Since then amine-based systems have been used to meet CO_2 product specifications in industries

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ranging from natural gas production to the food and beverage industry [32]. A number of vendors currently offer commercial amine-based processes, including the Fluor Daniel Econamine FG Plus process, the Mitsubishi Heavy Industries KM-CDR process, the Lummus Kerr-McGee process, the Aker Clean Carbon Just Catch process, the Cansolv CO_2 capture system and the HTC Purenergy Process [33].

The hundreds of commercial aqueous amine systems currently in operation typically vent the captured CO_2 to the atmosphere. Table 4 lists three recent projects at natural gas treatment plants (two in Norway, one in Algeria), in which the captured CO_2 is sequestered in deep geological formations to prevent its release to the atmosphere. The Statoil natural gas production facility at Sleipner in the North Sea, has been operating since 1996 and is the longest-running commercial CCS project. Fig. 13 shows the amine-based capture unit installed more recently at a natural gas treatment plant in Algeria. That unit is part of an integrated CCS system that includes CO_2 capture, pipeline transport and sequestration in a depleted gas formation.

As shown in Table 4, CO_2 is also captured at several coal-fired and gas-fired power plants where a portion of the flue gas stream is fitted with a CO_2 capture system. Fig. 14 shows the amine systems installed at two U.S. power plants, one burning coal, and the other natural gas. Here, the captured CO_2 is sold to nearby food processing facilities, which use it to make dry ice or carbonated beverages. However, these products soon release the CO_2 to the atmosphere, so there is no long-term sequestration.

To date, only ABB Lummus (now CB&I Lummus) has commercial flue gas CO_2 capture units operating at coal-fired power plants, while both Fluor Daniel and MHI have commercial installations at gas-fired plants (see Table 4). Both Fluor and MHI now also offer commercial guarantees for post-combustion capture at coal-fired power plants. In most cases the exact composition of commercial amine-based solvents is proprietary. The currently operating Lummus systems employ a solution of 20 percent MEA in water, while the Fluor systems use a solvent with a 30 percent amine [34,35]. Higher amine concentrations are beneficial in reducing the energy penalty of CO_2 capture since there is less water in the solution that has to be pumped and heated in the regeneration process. Capital cost also is reduced since higher amine concentrations lead to smaller equipment sizes. On the other hand, amines such as MEA are highly corrosive, so higher concentrations require chemical additives or more costly materials of construction to prevent corrosion. Tradeoffs among these factors underlie some of the differences in capture system designs offered by different vendors.

4.1.2. *Full-scale demonstration plants*

Although several CO_2 capture systems have operated commercially for nearly two decades on a portion of power plant flue gases, no capture units have yet been applied to the full flue gas stream of a modern coal-fired or gas-fired power plant. Thus, the operability and reliability of plant operation have yet to be demonstrated under conditions including larger equipment sizes and full integration of the capture unit and plant steam cycle. One or more demonstrations of post-combustion CO_2 capture at full-scale are thus widely regarded as crucial for gaining acceptance of this technology by electric utility companies, as well as by the institutions that finance and regulate power plant construction and operation. The European Union has called for twelve such demonstrations in Europe, while U.S. studies have called for at least six to ten full-scale projects to demonstrate a suite of capture technologies [13,36,37].

A major impediment has been the high cost of large-scale projects: roughly one billion dollars for CO_2 capture at a 400 MW unit operating for five years [37]. Several previously announced demonstrations of full-scale power plant capture and storage systems were delayed or canceled due to sharp escalations in construction costs prior to 2008, including a 160 MW demonstration project in the U.S. that was canceled not long after being announced [38]. More recently, two additional projects, one in the U.S. the other in the U.K., also were canceled because of cost considerations.

Table 5 lists the features and locations of the major post-combustion capture demonstration projects planned at power plants in the United States and other countries as of September 2011. Most of these CO_2 capture systems would be installed at existing coal-fired plants, with the captured CO_2 transported via pipeline to a geological storage site, often in conjunction with enhanced oil recovery to reduce project costs.

Note that while most of the projects in Table 5 plan to employ amine-based capture systems, some propose to use an ammonia-based process. Two such processes currently at the pilot plant stage are described in more detail below. Note too that most of the planned demonstration projects have expected startup dates of 2014 or later. This means that such projects are currently in the early stages of detailed design and that final commitments of full funding for construction have not yet been made. Similarly, it is still too early to know the details of capture system designs and the extent to which they can be expected to achieve improvements in CO_2 capture efficiency and/or reductions in cost relative to current commercial systems.

Table 4
Commercial post-combustion capture processes at power plants and selected industrial facilities that capture, transport and sequester CO_2 in an integrated CCS system [28–31].

Project name and location	Plant and fuel type	Year of startup	Approx. capture plant capacity	Capture system type (vendor)	CO_2 captured (10^6 tonnes/yr)
Projects in the U.S.					
IMC Global Inc. Soda Ash Plant (Trona, CA)	Coal and petroleum coke-fired boilers	1978	43 MW	Amine (Lummus)	0.29
AES Shady Point Power Plant (Panama City, OK)	Coal-fired power plant	1991	9 MW	Amine (Lummus)	0.06
Bellingham Cogeneration Facility (Bellingham, MA)	Natural gas-fired power plant	1991	17 MW	Amine (Fluor)	0.11
Warrior Run Power Plant (Cumberland, MD)	Coal-fired power plant	2000	8 MW	Amine (Lummus)	0.05
Projects Outside the U.S.					
Soda Ash Botswana Sua Pan Plant (Botswana)	Coal-fired power plant	1991	17 MW	Amine (Lummus)	0.11
Sumitomo Chemicals Plant (Japan)	Gas & coal boilers	1994	8 MW	Amine (Fluor)	0.05
Statoil Sleipner West Gas Field (North Sea, Norway)	Natural gas separation	1996	N/A	Amine (Aker)	1.0
Petronas Gas Processing Plant (Kuala Lumpur, Malaysia)	Natural gas-fired power plant	1999	10 MW	Amine (MHI)	0.07
BP Gas Processing Plant (In Salah, Algeria)	Natural gas separation	2004	N/A	Amine (Multiple)	1.0
Mitsubishi Chemical Kurosaki Plant (Kurosaki, Japan)	Natural gas-fired power plant	2005	18 MW	Amine (MHI)	0.12
Snøhvit Field LNG and CO_2 Storage Project (North Sea, Norway)	Natural gas separation	2008	N/A	Amine (Aker)	0.7
Huaneng Co-Generation Power Plant (Beijing, China)	Coal-fired power plant	2008	0.5 MW	Amine (Huaneng)	0.003

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Fig. 13. An amine-based CO_2 capture systems used to purify natural gas at BP's In Salah plant in Algeria; Photo courtesy of IEA Greenhouse Gas Programme.

4.1.3. *Pilot plant projects*

Table 6 lists a number of pilot-scale post-combustion CO_2 capture projects that are currently operating, or are in the design or construction stage, or have recently been completed. Most of these projects are testing and developing new or improved amine-based solvents with other projects testing ammonia-based solvents and calcium-bases sorbents. Planned pilot projects also include testing of capture processes based on concentrated piperazine, amino acid salts, solid sorbents and membrane-based systems.

4.1.3.1. Amine-based capture processes. The class of solvents called amines (more properly, alkanolamines) are a family of organic compounds that are derivatives of alkanols (commonly called the alcohols group) that contain an "amino" (NH_2) group in its chemical structure. Because of this complexity, there are multiple classifications of amines, each of which has different characteristics relevant to CO_2 capture [39]. For example, MEA reacts strongly with acid gases like CO_2 and has a fast reaction time and an ability to remove high percentages of CO_2, even at the low CO_2 concentrations found in flue gas streams. Other properties of MEA, however, are undesirable, such as its high corrosivity and regeneration energy requirement. Research groups are involved in synthesizing and testing a variety of amine mixtures and "designer" amines to achieve a more desirable set of overall properties for use in CO_2 capture systems. One major focus is on lowering the energy required for solvent regeneration to reduce process cost. Often, however, there are complex tradeoffs to consider [40]. High costs of manufacturing a new solvent also may detract from its thermochemical benefits. Pilot plant projects are acquiring the data needed to assess such tradeoffs and optimize an overall process.

4.1.3.2. Ammonia-based capture processes. A 2005 study by DOE/NETL found that post-combustion CO_2 capture using ammonia appeared promising, in part because ammonia is inexpensive, but also because it potentially could operate with a much smaller energy penalty than amines. The report suggested that if a number of engineering challenges could be overcome, the overall cost of an ammonia-based system would be substantially less than an amine-based system for CO_2 capture. Since ammonia potentially could capture multiple pollutants simultaneously (including CO_2, SO_2,

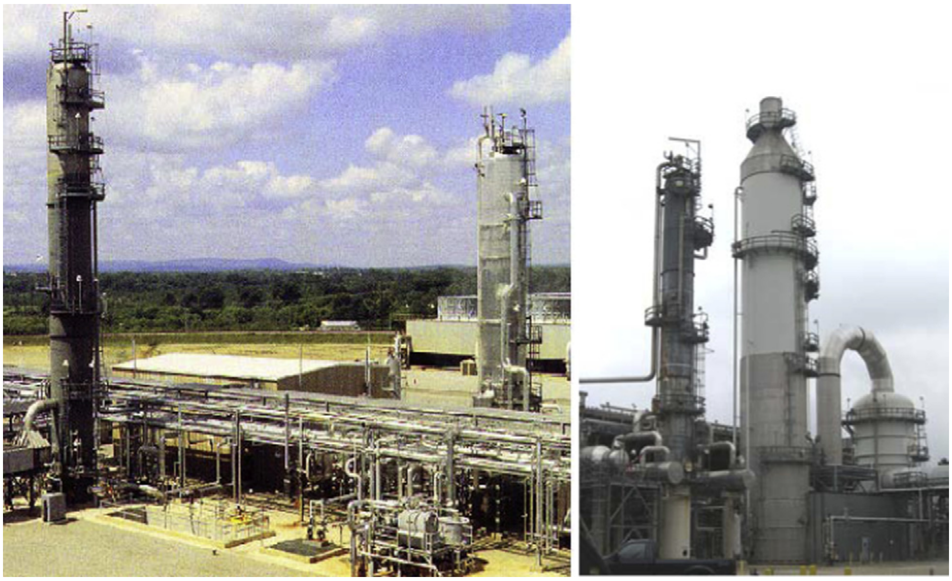

Fig. 14. Amine-based post-combustion CO_2 capture systems treating a portion of the flue gas from a coal-fired power plant in Oklahoma, USA (left) and a natural gas combined cycle (NGCC) plant in Massachusetts, USA (right); Photos courtesy of ABB Lummus, Fluor Daniels and Chevron.

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Table 5
Planned demonstration projects at power plants with post-combustion capture [28–31].

Project name and location	Plant and fuel type	Planned year of startup	Approx. capture plant capacity	Capture system type (vendor)	Annual CO_2 captured (10^6 tonnes)
Projects in the U.S.					
TenaskaTrailblazer Energy Center (Sweetwater, TX)	Coal-fired power plant	2014	600 MW	Amine (Fluor)	4.3
NRG Energy WA Parish Plant (Houston, TX)	Coal-fired power plant	2015	240 MW	Amine (Fluor)	1.5
Projects outside the U.S.					
SaskPower Boundary Dam Polygon (Estevan, Canada)	Coal-fired power plant	2014	115 MW	Amine (Cansolv)	1.0
TransAlta Project Pioneer Keephills 3 Power Plant (Wabamun, Canada)[a]	Coal-fired power plant	2015	200 MW	Chilled Ammonia (Alstom)	1.0
Vattenfall Janschwalde (Janschwalde, Germany)[a]	Coal-fired power plant	2015	125 MW	Amine (TBD)	N/A
PGE Bechatów Power Station (Bechatów, Poland)	Coal-fired power plant	2015	360 MW	Amine (Alstom, Dow Chemical)	1.8
Porto Tolle (Rovigo, Italy)	Coal-fired power plant	2015	200 MW[b]	Amine (TBD)	1.0
SSE Peterhead Power Station (Peterhead, UK)	Gas-fired power plant	2015	385 MW	N/A	1.0

N/A = not available; TBD = to be determined.

[a] These projects were recently cancelled during the final production of this paper.

[b] Estimated from other reported data.

NO_x and Hg), the overall plant cost could be reduced even further [41]. These considerations led to early estimates that the overall energy penalty of an ammonia-based system could be reduced to about half that of a conventional amine system—claims not substantiated in subsequent testing. Ammonia also has a higher volatility than MEA and thus is more easily released into the flue gas stream during the absorption step. Controlling this “ammonia slip” to acceptable levels is one of the major engineering challenges since any need for subsequent cleanup would add considerably to the cost [42]. The development of ammonia-based capture technology has advanced to the pilot plant stage and a commercial-sized demonstration has been planned.

In the chilled ammonia process developed by Alstom, the flue gas and CO_2 absorber are cooled to about 20 °C (68 °F) to reduce ammonia slip. In the absorber, ammonium carbonate is used to capture the CO_2. As with amine systems, the CO_2-rich stream is then sent to a regenerator where steam extracted from the power plant steam turbine is used to strip CO_2 from the solution. The CO_2-lean stream is then recirculated back to the absorber, as depicted in Fig. 15. Alstom has operated two chilled ammonia pilot plants—one in the United States (see Fig. 15) and one in Norway (see Table 6). The pilot plant at the Mountaineer power station in West Virginia (now closed) captured CO_2 from a flue gas slip stream equivalent to about 20 MW. This was the first successful integration of CO_2 capture, transport and geological sequestration at a coal-fired power plant.

In 2005, the Powerspan ECO process—which uses ammonia to capture SO_2 and NO_x from power plant flue gas streams—was modified to also capture CO_2. This process, called ECO_2, is similar to the chilled ammonia process in that it also uses ammonium carbonate to capture CO_2, though at a higher temperature. Ammonium sulfate from the SO_2 capture step is used to control ammonia slip so that ammonia is not consumed in the process. Thus, while amine-based systems must severely limit exposure of the solvent to acid gases like SO_2 and NO_2 to prevent solvent loss and degradation, ammonia does not degrade in the presence of these gases; instead, it forms ammonium sulfate and nitrate, which have value as fertilizer by-products [33,43]. Powerspan is testing its

Table 6
Pilot plant processes and projects post-combustion CO_2 capture [28–31].

Project name and location	Plant and fuel type	Planned year of startup	Approx. capture plant capacity	Capture system type (vendor)	Annual CO_2 captured 10^6 tonnes)
Projects in the U.S.					
First Energy R.E. Burger Plant (Shadyside, OH)	Coal-fired power plant	2008	1 MW	Ammonia (Powerspan)	0.007
American Electric Power Mountaineer Plant (WV)	Coal-fired power plant	2009	20 MW	Chilled Ammonia (Alstom)	0.1
Dow Chemicals, South Charleston Plant (WV)	Coal-fired power plant	2009	0.5 MW[a]	Amines (Dow/Alstom)	0.002
Projects Outside the U.S.					
Nanko Natural Gas Pilot Plant (Osaka, Japan)	Gas-fired power plant	1991	0.1 MW	Amine (MHI)	0.001
Matsushima Coal Plant (Nagasaki, Japan)	Coal-fired power plant	2006	0.8 MW[a]	Amine (MHI)	0.004
Munmorah Pilot Plant (Lake Munmorah, Australia)	Coal-fired power plant	2008	1 MW[a]	Ammonia (Delta, CSIRO)	0.005
Tarong Power Station (Nanango, Australia)	Coal-fired power plant	2008	0.5 MW[a]	Amine (Tarong & CSIRO)	0.0015
Hazelwood Carbon Capture (Morewell, Australia)	Coal-fired power plant	2008	2 MW	Amine (Process Group)	0.01
CASTOR CO_2 Capture to Storage (Esbjerg, Denmark)	Coal-fired power plant	2008	3 MW	Amine (Multiple)	0.008
Eni and Enel Federico II Brindisi Power Plant (Cortemaggiore, Italy)	Coal-fired power plant	2009	1.5 MW	Amine (Enel)	0.008
CATO-2 CO_2 Catcher (Rotterdam, Netherlands)	Coal-fired power plant	2008	0.4 MW	Amine (Multiple)	0.002
Limestone-Based Absorption of CO_2 (LISA) (Darmstadt, Germany)	Coal-fired power plant	2010	1 MW_{th} (~0.3 MW)	Carbonate looping	N/A
CaOling project (Mieres, Spain)	Coal-fired power plant	2011	1.7 MW_{th} (~0.6 MW)	Carbonate looping	0.007
Statoil Mongstad Cogeneration Pilot (Mongstad, Norway)	Natural gas-fired power plant	2012	15 MW[a] 7 MW[a]	Chilled NH_3 (Alstom) Amine (Various)	0.08 0.02
PGE Bechatów Power Station (Bechatów, Poland)	Coal-fired power plant	2014	20 MW	Amine (Alstom, Dow Chemical)	0.1

N/A = not available.

[a] Estimated from other reported data.

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ECO$_2$ process at a 1 MW pilot plant at First Energy's R.E. Burger plant, as indicated in Table 6.

4.1.3.3. Calcium-based capture processes. Some new post-combustion capture processes employ solid sorbents rather than liquid solvents to capture CO$_2$ from the flue gas of a coal-fired power plant. An advanced concept called the calcium looping cycle or carbonate looping cycle is currently at the small pilot plant stage, as noted in Table 6. In this process the desulfurized flue gas passes through a carbonator bed where calcium oxide (CaO) reacts with CO$_2$ in the flue gas to form calcium carbonate (CaCO$_3$). The carbonate is then heated in a separate reactor called the calciner where the reverse reaction takes place, releasing the CO$_2$. Energy for this reaction is supplied by combusting a fuel such as coal with high-purity oxygen. The CaO formed in the calciner is then sent back to the carbonator to complete the loop [44]. A schematic of this process is shown in Fig. 16. Fluidized bed reactors are likely to be the technology of choice for this process design since they have been proven for large-scale solids handling in other industrial applications. Studies indicate that calcium looping has the potential to be more efficient and lower in cost than amine-based capture methods [45,46], though as with other solid sorbent systems challenges remain, as discussed below in Section 4.1.4.

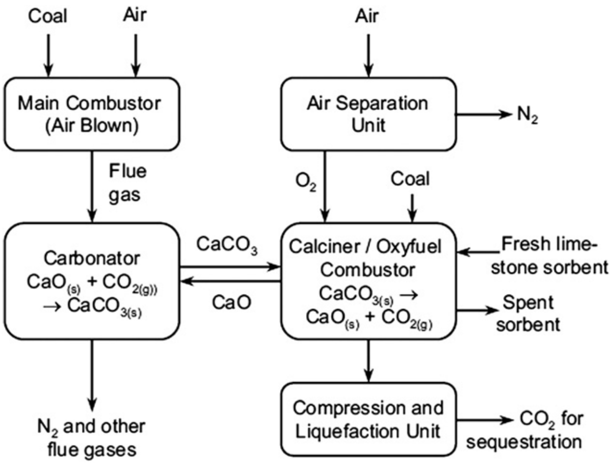

Fig. 16. Schematic of a calcium looping cycle [44].

4.1.4. Laboratory or bench-scale processes

A large number of new processes and materials for post-combustion CO$_2$ capture are currently at the laboratory or bench-scale stage of development [43]. These can be grouped into three general categories: (1) liquid solvents (absorbents) that capture CO$_2$ via chemical or physical mechanisms; (2) solid adsorbents that capture CO$_2$ via physical or chemical mechanisms; and (3) membranes that selectively separate CO$_2$ from other gaseous species. Within each category a number of approaches are being pursued, as summarized in Table 7.

Each of the approaches in Table 7 has some potential to reduce the cost and/or improve the efficiency of CO$_2$ capture relative to current commercial systems. At this early stage of development, however, it is difficult or impossible to reliably quantify the potential benefits or the likelihood of success in advancing to a commercial process. Indeed, at this stage many of the approaches being investigated consist solely of a novel or advanced material that holds promise for CO$_2$ capture, but which remains to be developed into an engineered process that can properly be called a capture technology. Thus, even if a new material succeeds in capturing CO$_2$ more efficiently or with a lower energy penalty, substantial challenges remain in incorporating such materials into a viable and scalable technology that is more economical than current CO$_2$ capture systems [47]. Thus, while some of the approaches in Table 7 may later advance to pilot-scale testing, others may not move past the bench-scale. The sections below describe in greater detail the promise and challenges for each of these options.

4.1.4.1. Liquid solvent-based approaches. Liquid solvents (typically a mixture of a base and water) selectively absorb CO$_2$ through direct contact between the chemical solvent and the flue gas stream. Regeneration of the solvent and release of CO$_2$ then takes place in a separate vessel (the regenerator or stripper) through a change of process conditions, such as a swing in temperature or pressure.

In general, the aim of solvent research is to identify or create new solvents or solvent mixtures that have more desirable characteristics than currently available solvents. Such properties include increases in CO$_2$ loading, reaction rates, thermal stability

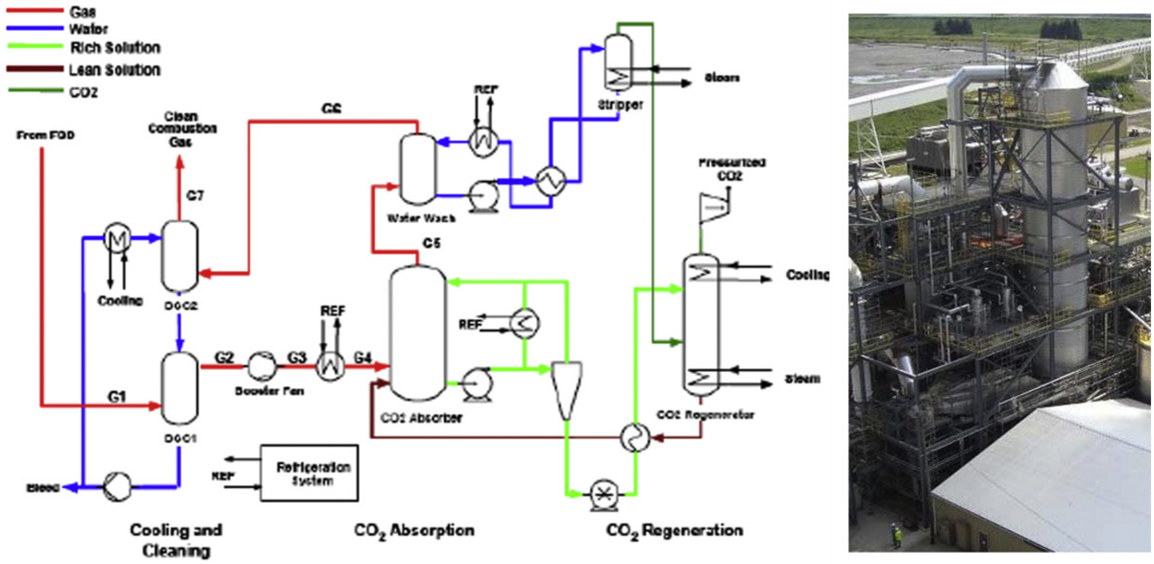

Fig. 15. Schematic of the chilled ammonia process for CO$_2$ capture (left) and the 20 MW pilot plant at the AEP Mountaineer station in West Virginia (right); Photo courtesy of AEP.

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Table 7
Post-combustion capture approaches being developed at the laboratory or bench-scale.

Liquid solvents	Solid adsorbents	Membranes
Advanced amines	Supported amines	Polymeric
Potassium carbonate	Carbon-based	Amine-doped
Advanced mixtures	Sodium carbonate	Integrated with absorption
Ionic liquids	Crystalline materials	Biomimetic-based

and oxidative stability, along with decreases in regeneration energy, corrosivity, viscosity, volatility and chemical reactivity with flue gas impurities. All of these attributes tend to lower the cost of CO_2 capture compared to current solvents.

Unfortunately, most real solvents exhibit a combination of desirable and undesirable properties. The latter include not only thermodynamic properties but also issues such as corrosivity and toxicity. Laboratory and bench-scale research thus seek new solvents that yield a more optimal blend of properties while meeting other requirements for system operability and safety. Table 8 summarizes the main advantages and challenges associated with advanced liquid solvent-based approaches to post-combustion CO_2 capture.

Examples of promising solvents include new amine formulations, carbonates, certain blends of amines and carbonates, and ionic liquids. One of the promising new amines receiving attention is piperazine. This solvent, currently being studied at the University of Texas and elsewhere, has been shown to have faster kinetics, lower thermal degradation and lower regeneration energy requirements than MEA in experiments thus far [32,49]. Further characterization studies are in progress to determine whether a blend of piperazine and other amines can offer improved performance for post-capture systems.

Potassium carbonate solvents, which have been used successfully in other gas purification applications, are now being investigated for bulk CO_2 capture from flue gases [50–52]. Potassium carbonate absorbs CO_2 through a relatively low-energy reaction, but has slow kinetics. Researchers are attempting to speed up absorption by blending potassium carbonate with various amines, with promising results [48,53]. Modeling of piperazine-promoted blends, for example, has suggested that due to improved kinetics and low regeneration energy requirements, such systems could have smaller equipment sizes and would be less energy-intensive than MEA-based systems [54].

Ionic liquids are liquid salts with low vapor pressure (hence, low solvent losses) that potentially can absorb CO_2 at high temperatures with relatively low regeneration energy requirements [31,48]. Researchers at the University of Notre Dame have shown that ionic liquids can capture SO_2 as well as CO_2, leading to the possibility that they can be used in a multi-pollutant capture system [42]. Georgia Tech Research Corporation is developing a different class of solvents called reversible ionic liquids which chemically react with CO_2 to make other ionic liquids which further absorb CO_2 [55]. One challenge for ionic liquids is that they can become highly viscous when absorbing CO_2, thus increasing the energy required for solvent pumping and the potential for mass transfer problems and operational difficulties in engineered processes [32,43]. The high cost of ionic liquids is another factor that could influence its competitiveness with other solvents.

Table 8
Technical advantages and challenges for advanced post-combustion solvents [48].

Description	Advantages	Challenges
Solvent reacts reversibly with CO_2, often forming a salt. The solvent is regenerated by heating (temperature swing), which reverses the absorption reaction (normally exothermic). Solvent is often alkaline.	Chemical solvents provide fast kinetics to allow capture from streams with low CO_2 partial pressure. Wet-scrubbing allows good heat integration and ease of heat management (useful for exothermic absorption reactions)	The large amount of steam required for solvent regeneration de-rates the power plant significantly. Energy required to heat, cool and pump non-reactive carrier liquid (usually water) is often significant. Vacuum stripping can reduce regeneration steam requirements but is expensive; bad economy of scale. Multiple stages and recycle stream may be required.

4.1.4.2. Solid sorbent-based approaches. Solid sorbents capture (adsorb) CO_2 on their surfaces, as illustrated in Fig. 17. They then release the CO_2 through a subsequent temperature or pressure change, thus regenerating the original sorbent. Solid sorbents have the potential for significant energy savings over liquid solvents, in part because they avoid the need for the large quantities of water that must be repeatedly heated and cooled to regenerate the solvent solution [42,56]. This reduces the cost of regeneration. Sorbent materials also have lower heat capacity than solvents and thus require less regeneration energy to change their temperature.

There are challenges, however, in how to efficiently get heat into and out of a solid sorbent material. More complicated solids handling equipment also is required compared to solvent solutions, though experience with mature industrial technologies such as catalytic crackers indicates that large-scale solids handling is certainly achievable. Resistance to physical attrition and deterioration over time is another important property for most solid sorbent applications. Finally, it is not yet clear which of several different absorber designs for solid sorbents (e.g., fluidized beds, packed bed reactors, transport reactors, or other systems) will be most effective in reducing overall cost in a particular application.

In general, the aim of solid sorbent research is to reduce the cost of CO_2 capture by designing durable sorbents with efficient materials handling schemes, increased CO_2 carrying capacity, lower regeneration energy requirements, faster reaction rates and minimum pressure drops [48]. The CO_2 carrying capacity is a key sorbent parameter that depends on the total microscopic surface area of the material. Researchers are thus attempting to identify and design sorbents with very high surface area for CO_2 capture [31]. The capture mechanism can be either a chemical or physical surface interaction. Solid sorbents that rely on chemical mechanisms are similar to liquid solvents. They include amines supported on the surface of other materials (called supported amines), as well as carbonates such as calcium carbonate (limestone) and sodium carbonate (soda ash). Sorbents that rely on physical surface interactions include materials such as activated carbon, zeolites and metal organic frameworks (MOFs).

Supported amines share the benefits of liquid amine solvents but require less energy to regenerate because there is no water solution [57]. The amine sorbent can be physically supported by a number of different materials, including relatively inexpensive activated carbon [58]. Such sorbents have been shown to have high CO_2 carrying capacities compared to many other solid sorbents [59]. Current research is focused on issues of thermal stability and fouling, as these sorbents have a tendency to break down over time and degrade in the presence of SO_2 [55].

Sodium carbonate-based sorbents have been recognized for their CO_2 capture potential, although their performance is degraded by contaminants in flue gas [31,60]. Another promising development is a system using a sodium carbonate-based sorbent for CO_2 capture at coal or gas-fired power plants [42].

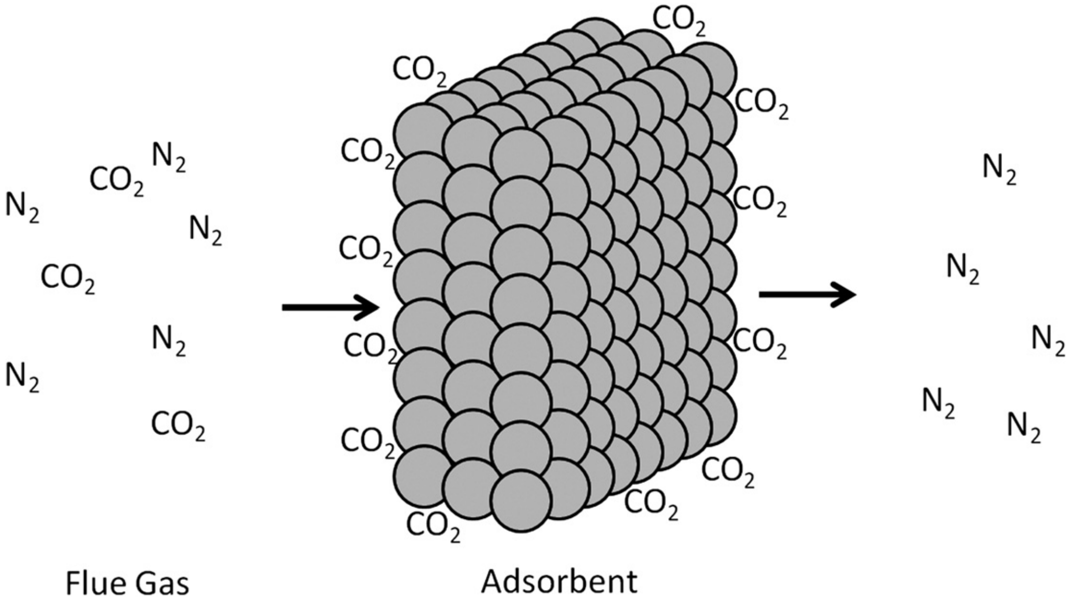

Fig. 17. Schematic of CO_2 adsorption on the surfaces of a solid sorbent (The simplified flue gas composition is represented as a mixture of CO_2 and nitrogen, N_2, the principal flue gas constituent).

Carbon-based adsorbents such as activated carbon and charcoal also are attractive because they are relatively inexpensive and have large surface areas that can readily adsorb CO_2. Researchers at the University of Wyoming, for example, claim that their Carbon Filter Process potentially can capture 90 percent of flue gas CO_2 and regenerate it with at least 90 percent CO_2 purity at a lower-cost than amine-based processes [47,61]. Carbon also can provide a support material for amines or other solid sorbents.

Metal organic frameworks (MOFs) and zeolites are crystalline sorbents that are also receiving attention for post-combustion CO_2 capture. MOFs consist of a matrix structure of metallic and organic molecules containing void spaces that potentially can be used to absorb large amounts of CO_2 with low regeneration energy requirements and cost. Zeolites are porous alumino-silicate materials that have high selectivity, but low carrying capacity for CO_2 and are subject to performance degradation in the presence of water [33,43]. Researchers at the University of Akron are investigating an approach combining zeolites with amines to improve overall performance [42].

Table 9 summarizes the key advantages and challenges of solid sorbent-based approaches to post-combustion CO_2 capture. Though such systems have the potential to offer better performance than current amine systems, the need to handle large amounts of solids tends to make this approach more complex and more difficult to scale-up than an equivalent liquid solvent system. Sorbents also must have high selectivity for CO_2 and be relatively insensitive to trace impurities in the flue gas. Because CO_2 bonding to sorbents is not as strong as with chemical interactions, multiple contacting stages also may be required to achieve high CO_2 capture efficiencies, which would increase process costs [31, 33, and 43]. Current R&D programs are attempting to address these challenges.

4.1.4.3. Membrane-based approaches. Membranes are permeable materials that can be used to selectively separate CO_2 from other components of a gas stream. They effectively act as a filter, allowing only (or mostly) CO_2 to pass through the material. The driving force for this separation process is a pressure differential across the membrane, which can be created either by compressing the gas on the feed side of the material or by creating a vacuum on the downstream side.

Membranes have been used for gas purification in a number of industrial applications since the 1980s [62,63]. Two important physical parameters of a membrane are its selectivity and permeability. Selectivity reflects the extent to which a membrane allows some molecules to be transported across the material, but not others. For post-combustion CO_2 capture, the selectivity to CO_2 over N_2 (the main constituent of flue gas) determines the purity of the captured CO_2 stream. The permeability of a membrane reflects the amount of a given substance that can be transported for a given pressure difference [64]. This determines the membrane surface area needed to separate and capture a given amount of CO_2. Ideally,

Table 9
Technical advantages and challenges for solid sorbent approaches to post-combustion CO_2 capture [48].

Description	Advantages	Challenges
When sorbent pellets are contacted with flue gas, CO_2 is absorbed onto chemically reactive sites on the pellet. Pellets are then regenerated by a temperature swing, which reverses the absorption reaction.	Chemical sites provide large capacities and fast kinetics, enabling capture from streams with low CO_2 partial pressure. Higher capacities on a per mass or volume basis than similar wet-scrubbing chemicals. Lower heating requirements than wet-scrubbing in many cases (CO_2 and heat capacity dependent).	Heat required to reverse chemical reaction (although generally less than for wet-scrubbing). Heat management in solid systems is difficult. This can limit capacity and/or create operational issues for exothermic absorption reactions. Pressure drop can be large in flue gas applications. Sorbent attrition may be high.

a combination of high selectivity and high permeability for CO_2 is desired in a membrane for CO_2 capture.

Among the current laboratory and bench-scale developments in this area, researchers at the University of Mexico are attempting to incorporate amine functional groups into membrane materials—a development that could help raise the selectivity of CO_2 [33,42,43]. Another active research area is gas absorption membranes [47]. Here, CO_2-laden flue gases contact one side of a membrane while a liquid solvent (such as an amine-based solvent) contacts the other side. As CO_2 and other gases pass through the membrane, the CO_2 is selectively absorbed by the liquid solvent [31]. This approach holds potential for better performance than conventional absorber and stripper configurations [43].

Yet another approach employs membranes with biomimetic components, seeking to employ processes found in nature. One such process uses the enzyme carbonic anhydrase, which facilitates the transport of CO_2 in the respiratory system of mammals [65]. One effort to exploit this process is a liquid membrane system catalyzed by carbonic anhydrase [42]. While preliminary results show potential for significant decreases in energy penalty and cost compared to amine-based systems, the significant challenges that remain include the problems of membrane fouling and scale-up to power plant applications.

Table 10 summarizes the potential benefits and technical challenges of membrane-based technologies for post-combustion CO_2 capture. By most accounts, membranes today are not economically competitive with amine-based post-combustion CO_2 capture [43]. Challenges include the need for high selectivity, large surface areas to process power plant flue gases, limited temperature ranges for operation, low tolerance to flue gas impurities (or requirements for additional equipment to remove those impurities) and high parasitic energy requirements to create a pressure differential across the membrane [66].

Despite these issues, there are strong proponents of membranes for post-combustion CO_2 capture. For example, Favre (2007) asserts that many of the challenges for membrane technology are amenable to engineering solutions, such as the higher surface areas achievable with hollow filter membranes compared to spiral-wound designs. He also notes that membranes could be more competitive with amines in applications with higher CO_2 concentrations, such as in the cement and steel industries. A power plant boiler fired by oxygen-enriched air also would increase the CO_2 concentration of the flue gas, making membrane-based separation more competitive [63].

4.1.5. *Conceptual design stage*

This stage of process development typically involves engineering analyses or computer-based modeling studies of novel capture technology concepts or systems whose fundamental principles are usually well understood, but which are lacking the experimental data needed to test or verify the merits of the idea. Here we briefly discuss three classes of novel but untested approaches to carbon capture: novel sorbents, hybrid systems and novel regeneration methods.

4.1.5.1. Novel sorbents. A number of research groups are investigating the development of ultra-high surface area porous materials for CO_2 capture, including metal organic frameworks (discussed earlier), zeolytic imidizolate frameworks and porous organic polymers. These materials have pore sizes, surface areas and chemistries that are highly “tunable” so that molecules can, in principle, be designed and fabricated by chemists and materials scientists to maximize CO_2 capture performance. Because CO_2 capture research in this area is relatively new, very little work has yet been done to assess these materials under realistic capture conditions or to incorporate them into workable capture technologies.

4.1.5.2. Hybrid capture systems. Hybrid approaches to new solvents and sorbents attempt to combine the best features of two or more components to mitigate the undesirable properties of one component. For example, a typical problem with some CO_2 capture solvents is that they become highly viscous when interacting with CO_2. Hybrid approaches to solving this problem include supporting the solvent on either a membrane or a solid sorbent. In these cases, viscosity is no longer an issue since no liquids are flowing.

For solid sorbents, one of the key problems is how to get heat into the sorbent during regeneration, since heat transfer in gas-solid systems is not as efficient as in liquid systems. One proposed solution is to immobilize the sorbents on a membrane or other solid support material that allows heat to be transferred more efficiently between two solids in direct contact.

Some of these hybrid approaches have advanced to the laboratory or bench-scale, as noted earlier, while others are being studied at the conceptual stage. It is uncertain, however, how the cost of these systems will compare to that of a single-component system whose active capture agent is now “diluted” by the other component. In general, one expects that the capital cost will be higher for a hybrid system, so its CO_2 capture performance must be substantially improved to offset the higher cost.

4.1.5.3. Novel regeneration methods. The two most common ways of regenerating CO_2 capture solvents or sorbents is the application of heat (temperature swing) or a vacuum (pressure swing), both of which are energy-intensive and costly. Researchers are examining alternative approaches that could be more efficient and less costly.

One alternative (and theoretically more efficient) approach is based in electrochemistry. In these systems a flow of electrons is used to facilitate both the capture and regeneration steps. Of the several concepts that have been studied, the most promising applies electrochemistry to carbonate materials to make separate acid and base solutions (so-called pH swing systems), with one solution used as a solvent to capture CO_2 and the other used to regenerate the solvent [67]. This technology is similar to a fuel cell in that it requires electrodes and specialized membranes to

Table 10
Technical advantages and challenges for membrane-based post-combustion CO_2 capture [48].

Description	Advantages	Challenges
Uses permeable or semi-permeable materials that allow for the selective transport and separation of CO_2 from flue gas.	No steam load. No chemicals needed.	Membranes tend to be more suitable for high pressure processes such as IGCC. Tradeoff between recovery rate and product purity (difficulty to meet both at same time). Requires high selectivity (due to CO_2 concentration and low pressure ratio). Good pre-treatment. Poor economies of scale. Multiple stages and recycle streams may be required.

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selectively separate particular species, such as protons and hydroxide ions. Fig. 18 illustrates one of the conceptual designs.

There are two variations of the pH swing concept, electrolysis and electrodialysis. The energy required for electrolysis is high and similar to that required for electrolysis of water. However, besides capturing CO_2 the process also generates hydrogen and oxygen, which have additional economic value. Electrodialysis is a more efficient process, but no valuable gases such as hydrogen are produced. Electrodialysis has been used commercially to desalinate water, but is only just being studied for application to CO_2 capture [68]. A third electrochemical approach employs membranes to separate gases such as hydrogen, oxygen and CO_2. This approach is theoretically the most efficient, but high efficiencies have not been obtained in practice due to the limitations of existing materials [69]. While the fundamentals of electrochemical approaches to CO_2 capture have been proven at the bench-scale, complete process designs are still only conceptual at this time. Other concepts for regenerating CO_2 sorbents or solvents employ photochemical processes or electromagnetic radiation such as microwave heating [70]. However, it appears unlikely that such approaches will soon (if ever) move out of the conceptual stage because of either technical or economic limitations.

4.1.6. System studies

In addition to component-level studies of advanced CO_2 capture technologies, a variety of systems studies have been undertaken to analyze ways of improving the overall efficiency of power plants with CO_2 capture. One of the most promising methods is improved heat integration between the power plant and the CO_2 capture unit [17,71]. As noted in Section 2.2, measures that increase plant efficiency can reduce the cost of CO_2 capture, provided they do not introduce new costs that offset the efficiency benefits. For example, the need for additional heat exchangers may introduce costs that outweigh the gains from improved efficiency. Assessment of such measures typically requires the construction of fully integrated pilot plants or demonstration plants where the feasibility of such designs can be evaluated in greater detail.

4.1.7. Conclusion for post-combustion capture processes

While R&D activities to reduce the cost of post-combustion CO_2 capture have increased substantially in recent years, most current efforts are still at the early stages of technology development. This is seen clearly in Fig. 19, which shows the results of an Electric Power Research Institute (EPRI) study that reviewed over a hundred active projects in this field and ranked them on the TRL scale described earlier in Section 3 [72]. That study found that as of 2008 all but a few of the post-combustion capture projects were between TRLs 1 and 5, which corresponds to the conceptual design and laboratory/bench-scale categories used in this paper. Only a small number of projects were ranked at TRL 6, corresponding to the pilot plant stage. None were ranked at higher stages of development.

The EPRI study also shows that most of the new processes under development employ absorption methods (i.e., solvents) for post-combustion capture of CO_2. Fewer new processes and concepts utilize membranes or solid sorbents (adsorption) for CO_2 capture—a reflection of the greater challenges facing those approaches.

Key questions that remain are: What are the prospects for any of these projects to result in a viable new process for CO_2 capture? How much improvement in performance or reduction in cost can be expected relative to current or near-term options? How long will it take to see these improvements? Such questions are addressed later in Sections 7 and 8 following the discussions of pre-combustion and oxy-combustion CO_2 capture processes.

4.2. Status of pre-combustion capture processes

This section summarizes the status of current and emerging pre-combustion CO_2 capture technologies at various stages of development. Pre-combustion CO_2 capture can be used both in power plants and in other industrial processes where CO_2 separation is required, such as in synthetic fuels production. The more advanced capture systems include commercial chemical solvents used widely in natural gas and synthesis gas production. Processes at the earliest stages of development employ novel methods such as solid sorbents or membranes for CO_2 capture.

4.2.1. Commercial processes

Currently there are no commercial applications of pre-combustion CO_2 capture at electric power plants. Proposed IGCC power plant designs, however, would employ commercial processes such as Selexol™ or Rectisol® for CO_2 capture. Such processes are already widely used in other industrial applications

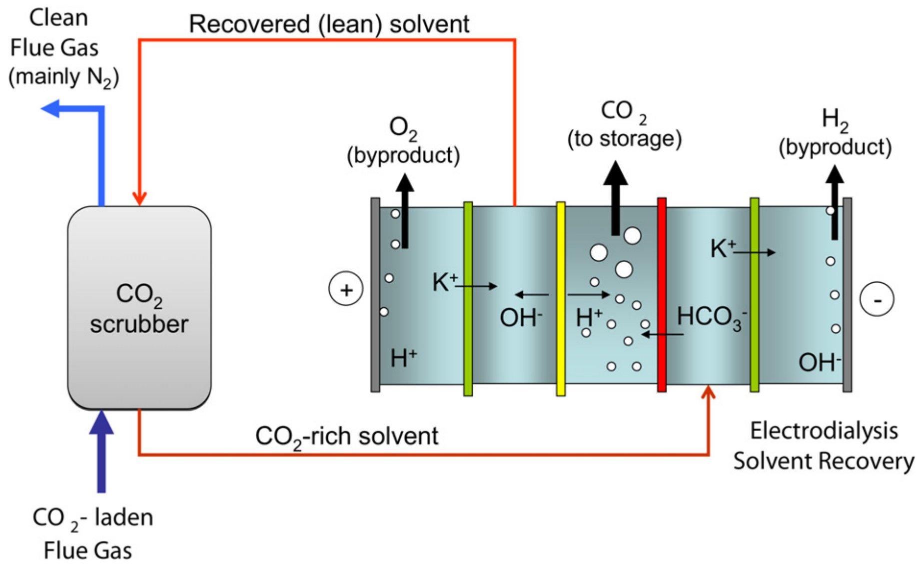

Fig. 18. Schematic of a process concept using electrodialysis to capture and regenerate CO_2, while generating hydrogen and oxygen as by-products [67].

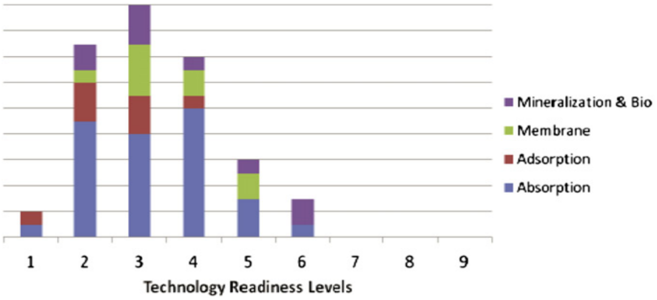

Fig. 19. Technical readiness levels (TRLs) of projects developing post-combustion capture using different approaches. The *y*-axis in this study was not scaled explicitly but corresponds to the relative number of processes of a given type. Also, the approach labeled "mineralization & bio" is considered in the present report to be a sequestration method rather than a post-combustion capture method since it requires a stream of concentrated CO_2 that has already been captured [72].

to remove contaminants such as sulfur and nitrogen compounds, as well as CO_2, present in syngas mixtures. Two examples are cited here to illustrate the scale at which pre-combustion capture technologies are currently used commercially.

The Farmlands chemical plant in Coffeyville, Kansas, shown in Fig. 20, uses the Selexol system to separate and capture CO_2 from a hydrogen-CO_2 gas mixture produced by the gasification of petroleum coke (petcoke) followed by a water-gas shift reactor—the same processes depicted earlier in Fig. 4 for an IGCC plant with pre-combustion CO_2 capture. At the Coffeyville plant, more than 93 percent of the CO_2 is captured, amounting to about 0.2 million tons of CO_2 per year [73]. A portion of this CO_2 is used to manufacture urea and the remainder is vented to the atmosphere. The separated stream of nearly-pure hydrogen is used to manufacture ammonia (rather than burned to generate electricity, as in an IGCC plant), with the ammonia subsequently used to produce fertilizers. This project has been in operation since 2000 and is similar to other industrial applications that use the Selexol process for CO_2 capture.

The Great Plains synfuels plant in North Dakota, operated by the Dakota Gasification Company and also shown in Fig. 20, employs coal gasification to produce synthetic natural gas. In that process, the plant captures approximately 3 million tons/year of CO_2 using the methanol-based Rectisol process. Previously, that CO_2 was vented to the atmosphere; now it is compressed and transported via a 205-mile pipeline to a Canadian oil field, where it is used for enhanced oil recovery and sequestered in the depleted oil reservoir. These two examples illustrate current commercial applications of pre-combustion CO_2 capture technologies that would be employed at gasification-based power plants. The choice of solvent or process would depend on the conditions of a particular project or application. The following section discusses current plans for full-scale demonstrations of pre-combustion capture at power plants.

4.2.2. *Full-scale demonstration plants*

As with post-combustion capture, to date there have been no full-scale demonstrations of pre-combustion CO_2 capture at an IGCC power plant, although a number of full-scale projects have been announced and one (in China) is currently under construction. Several other previously announced IGCC-CCS projects in different parts of the world have been delayed or canceled in recent years, including the FutureGen project originally slated for construction in Mattoon, Illinois [74]. Nevertheless, it appears reasonable that at least some of the large-scale projects currently planned for pre-combustion CO_2 capture in the U.S. and other countries will indeed materialize over the next several years, with costs shared between the public and private sectors.

Table 11 lists the features and locations of major announced demonstration of pre-combustion CO_2 capture. They include fuels production plants as well as IGCC power plants. Most of the projects in Table 11 would not begin operation until 2014 or later. In most cases the captured CO_2 would be sequestered in a depleted oil reservoir in conjunction with EOR. The percentage of CO_2 captured varies widely across these projects, from 50 to 90 percent of the carbon in the feedstock. Table 11 shows that Selexol is the preferred technology for pre-combustion capture at projects that have announced their selection. However, for most of the projects listed the choice of solvent or capture technology is not yet known.

Given the extensive commercial experience and scale of CO_2 capture in industrial processes with gas streams nearly identical to an IGCC plant, most of the large-scale projects in Table 11 will serve to demonstrate other aspects of IGCC technology. In particular, the reliability of gasifier operations and the large-scale use of hydrogen to power the gas turbine following CO_2 capture are key technical issues that remain to be demonstrated in the electric utility environment. The plant startup schedules in Table 11 indicate it will be at least a few years before significant operational data begins to accrue at most of the planned demonstration projects. As before, the possibility also remains that some of these planned projects may not materialize due to economic or other reasons.

4.2.3. *Pilot plant projects*

In general there is relatively little current development of pre-combustion CO_2 capture at the pilot plant scale. However, two projects at IGCC plants in Europe—Nuon's Buggenum plant in the

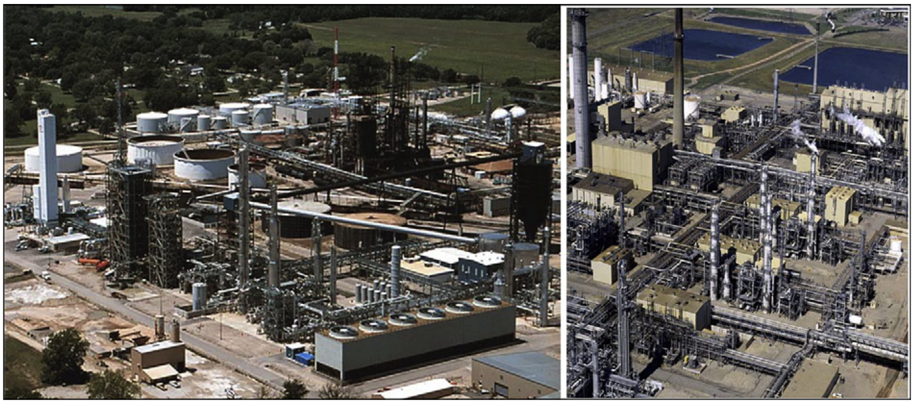

Fig. 20. A pre-combustion CO_2 capture system is used to produce hydrogen from gasified petcoke at the Farmlands plant in Kansas (left) and synthetic natural gas from coal at the Dakota gasification plant in North Dakota (right); Photos courtesy of UOP and IPCC.

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Table 11
Planned demonstration projects with full-scale pre-combustion capture [28–31].

Project name and location	Plant and fuel type	Planned year of startup	Plant size or capacity	CO_2 capture system	Annual CO_2 captured (10^6 tonnes)
Projects in the U.S.					
Baard Energy Clean Fuels (Wellsville, Ohio)	Coal + biomass-to-liquids	2013	53,000 barrels/day	Rectisol	N/A
DKRW Energy (Medicine Bow, WY)	Coal-to-liquids	2014	20,000 barrels/day	Selexol	N/A
SummitPower (Penwell, Texas)	Coal IGCC and polygen (urea)	2014	400 MW_g	Rectisol	3.0
Kemper County IGCC (KemperCounty, Miss.)	Lignite IGCC	2014	584 MW	N/A	~3
Wallula (Washington)	Coal IGCC	2014	600–700 MW	N/A	N/A
Taylorville Energy Center (Taylorville, Illinois)	Coal to SNG + IGCC	2014[a]	602 MW	N/A	3.0
Hydrogen Energy (Kern County, California)	Petcoke IGCC	2016	250 MW	N/A	2
Projects outside the U.S.					
GreenGen (Tianji Binhai, China)	Coal IGCC and poly-generation	2011 (stage I: no CCS)	250 MW	N/A	N/A
Eston Grange IGCC (Teesside, UK)	Coal IGCC	2012	800 MW	N/A	5
Dom Valley IGCC (Stainforth, UK)	Coal IGCC	2014	900 MW	Selexol	4.5
Genesee IGCC (Edmonton, Canada)	Coal IGCC	2015	270 MW	N/A	1.2
RWE Goldenbergwerk (Hurth, Germany)	Lignite IGCC	2015[b]	360 MW	N/A	2.3
Kedzierzyn Zero Emission Power and Chemicals (Opole, Poland)	Coal-biomass IGCC and polygen	2015	309 MW 500 ktons/yr methanol	N/A	2.4
Nuon Magnum (Eemshaven, Netherlands)	Multi-fuel IGCC	2015[c]	1200 MW_g	N/A	N/A
FuturGas (Kingston, Australia)	Lignite to liquids	2016	10,000 barrels/day	N/A	1.6

N/A = not available; MW_g = megawatts gross generated.

[a] This project is on hold pending future state funding.

[b] Depends on outcome of the Carbon Storage Law.

[c] Depends on performance of the Buggenum pilot plant (see Table 12).

Netherlands and Elcogas's Puertollano plant in Spain—are significant developments because they are the first applications of CO_2 capture at operating IGCC facilities, albeit at a small-scale treating only a portion of the syngas stream.

The Nuon Buggenum project is aimed at testing pre-combustion CO_2 capture in order to better select, design and optimize a capture system after some operating experience is gained. Both the water-gas shift reactors and the CO_2 capture process will be optimized for their performance efficiency and different physical and chemical solvents will be tested. The main aim of this pilot plant is to gain operational experience which can be used for the Nuon Magnum IGCC power plant listed earlier in Table 11 [75]. The ELCOGAS IGCC plant in Puertollano, Spain captured its first tonne of CO_2 in late 2010 [76]. Preliminary results from this pilot plant indicate operation as expected with final testing to be completed in mid-2011.

4.2.4. Laboratory or bench-scale developments

Though pre-combustion CO_2 capture has a lower energy penalty and lower-cost than post-combustion capture processes performing similar duty, there is scope for improvements that can further reduce costs. With this aim, current research is focused mainly on improving the capture efficiency so that the size and cost of equipment can be lowered. The same three approaches discussed in Section 4.1 for post-combustion capture are being pursued, namely, liquid solvents that separate CO_2 from a gas stream by absorption; solid sorbents that separate CO_2 by adsorption onto a solid surface; and membranes that separate CO_2 by selective permeation through thin layers of solid materials.

4.2.4.1. Solvent-based capture processes. As noted previously, current pre-combustion CO_2 capture systems employ solvents that selectively absorb CO_2 (and other acid gases) from a gas stream. Physical absorption is characterized by weak binding forces between gas molecules and the solvent molecules. Research on physical solvents is aimed at improving the CO_2 carrying capacity and reducing the heat of absorption. Higher carrying capacity means that more CO_2 is captured in every pass through the absorption tower, thus lowering costs. Solvents with a low heat of absorption require less energy to strip CO_2 during the regeneration step, which also lowers cost. Of the two properties, the main focus is on improving the CO_2 carrying capacity since the heat of absorption already is low for most physical solvents (which is why a pressure swing method can be used to strip captured CO_2 from the solvent, unlike chemical solvents where heat is needed).

The CO_2 carrying capacity of a solvent depends on a number of factors including certain properties of the solvent, the partial pressure of CO_2 in the gas stream and the temperature of the process. Usually, the carrying capacity increases at higher pressure and lower temperature. A practical problem with liquid solvents is their corrosivity. Any novel solvent must therefore have low corrosive properties. Table 13 summarizes the advantages of physical solvents and the challenges in improving their properties.

Research on new or improved solvents for pre-combustion capture seeks to develop solvents that allow CO_2 to be captured at higher pressures and temperatures. Currently, the syngas from a coal gasifier must be cooled to near room temperature before entering the solvent-based CO_2 capture unit. New solvents that can

Table 12
Pilot plant projects for pre-combustion CO_2 capture at IGCC power plants [28–31].

Project name and location	Plant and fuel type	Planned year of startup	Plant size or capacity	CO_2 capture system	Annual CO_2 captured (10^6 tonnes)
Nuon Buggenum (Buggenum, Netherlands)	Coal and biomass IGCC	2010	N/A	Different physical and chemical solvents	0.010
Elcogas Puertollano (Puertollano, Spain)	Coal and petcoke IGCC	2010	14 MW_{th} (~5 MW)	Different commercial solvents	0.035

MW_{th} = megawatts thermal.

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Table 13
Key advantages and challenges of physical solvents for pre-combustion CO_2 capture [48].

Description	Advantages	Challenges
Solvent readily dissolves CO_2. Solubility is directly proportional to CO_2 partial pressure and inversely proportional to temperature, making physical solvents more applicable to low-temperature, high pressure applications (cooled syngas). Regeneration normally occurs by pressure swing.	CO_2 recovery does not require heat to reverse a chemical reaction. Common for same solvent to have high H_2S solubility, allowing for combined CO_2/H_2S removal and disposal (where permitted). System concepts that recover CO_2 with some steam stripping rather than flashed, with delivery at a higher pressure, may optimize processes for power systems	CO_2 pressure is lost during flash recovery Must cool synthesis gas for CO_2 capture, then heat and humidify again for firing in gas turbine Low solubility can require circulating large volumes of solvent, which increases energy needs for pumping Some H_2 may be lost with the captured CO_2

capture CO_2 at higher temperatures can therefore increase overall plant efficiency and thus potentially reduce the equipment needs and cost of CO_2 capture. In this context, ionic liquids, discussed earlier in Section 4.1.5, are also being studied as potential solvents for CO_2 capture in pre-combustion applications [48]. Ionic liquids are salts that are liquid at room temperature. They have high CO_2 absorption potential and do not evaporate at temperatures as high as 250 °C. In an IGCC system, this could allow separation of CO_2 without cooling the syngas, thereby reducing equipment size and cost. This is also one of the approaches being pursued to develop new physical absorption solvents for pre-combustion capture [48].

4.2.4.2. Sorbent-based capture processes. Solid sorbents are another class of material that potentially could be used for pre-combustion CO_2 capture as well as for post-combustion capture (see Section 4.1). The primary advantage of solid sorbent systems over solvents in pre-combustion applications is their ability to operate at high temperatures. This avoids the additional equipment for syngas cooling, thus reducing cost. However, the handling of solids is generally more difficult than the handling of liquid-based systems. This offsets some of the advantages of solids and can be an important factor in the choice (and overall cost) between solvent and sorbent-based capture technology in large-scale applications.

Solid sorbent-based systems are used commercially today in a variety of applications, such as in hydrogen purification processes employing pressure swing adsorption. With some changes, that system has scope to be adapted to capture CO_2. LehighUniversity, RTI International, TDA Research, the University of North Dakota and the URS Group are among the organizations currently working on development of solid sorbents [48]. The work is primarily focused on identifying the most promising sorbent materials and conducting bench-scale experiments. Table 14 summarizes the key advantages and challenges of using solid sorbents for pre-combustion CO_2 capture.

4.2.4.3. Membrane-based capture processes. As described in Section 4.1.5, membrane-based capture processes operate by selectively allowing a gas to permeate through the membrane material. Membranes for CO_2 capture are made of micro-porous metallic, polymeric or ceramic materials. For effective CO_2 capture in pre-combustion applications, they must not only have high permeability and selectivity to CO_2, but also be able to operate at the high pressures and temperatures characteristic of IGCC systems.

Fig. 21 shows a schematic of a membrane separation process for CO_2 capture in an IGCC application, where CO_2 is preferentially separated from hydrogen in the gas stream following the water-gas shift and sulfur removal steps described earlier in Fig. 3. Because the separation is seldom perfect, several stages are typically needed to increase the purity of the separated components.

To date, membrane technology has been used commercially for gas purification and CO_2 removal in the production of hydrogen, but it has not been used specifically for pre-combustion CO_2 capture in IGCC plants or related industrial processes that require a high CO_2 recovery rate with high CO_2 purity. Applications to IGCC are of interest since the mixture of CO_2 and H_2 following the shift reactor is already at high pressure, unlike post-combustion applications which require additional energy to create a pressure differential across the membrane.

Table 15 summarizes the key advantages and challenges of membrane separation systems for pre-combustion capture applications. Many of the challenges discussed earlier for post-combustion applications also apply here. Because of their modular nature and the need for relatively large surface areas, membrane systems again do not have the economies of scale with plant size found in other types of capture systems. Thus, they must have substantially superior performance and/or lower unit cost to compensate. These are the major hurdles that current research is attempting to overcome.

4.2.4.4. Enhanced water-gas shift reactors. In an IGCC plant with CCS, the syngas exiting the gasifier is subjected to a water-gas shift (WGS) reaction to increase the concentration of CO_2 in the gas stream prior to CO_2 capture. This step also provides additional hydrogen (H_2) for power generation. The WGS reaction between carbon monoxide (CO) in the syngas and steam (H_2O) that is added is:

$$CO + H_2O \Leftrightarrow CO_2 + H_2$$

The thermodynamics of chemical reactions dictates that the speed and efficiency of this reaction is limited by the presence of

Table 14
Key advantages and challenges of solid sorbents for pre-combustion CO_2 capture [48].

Description	Advantages	Challenges
When sorbent pellets are contacted with syngas, CO_2 is physically adsorbed onto sites and/or dissolves into the pore structure of the solid. Rate and capacity are directly proportional to CO_2 partial pressure, making these sorbents more applicable to high pressure applications. Regeneration normally occurs by pressure swing.	CO_2 recovery does not require heat to reverse a reaction. Common for H_2S to also have high solubility in the same sorbent, so CO_2 and H_2S capture and disposal can be combined (where permitted). System concepts in which CO_2 is recovered with steam stripping rather than flashed and delivered at a higher pressure, which may optimize processes for power systems.	Solids handling is more difficult than liquid-gas systems. CO_2 capture with sorbents is a novel concept though other gas purification processes use adsorption techniques.

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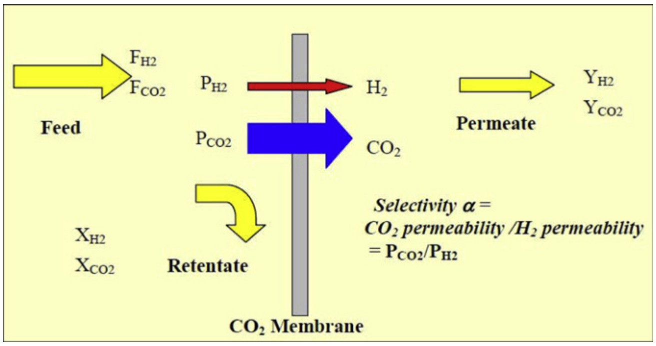

Fig. 21. Schematic of pre-combustion CO_2 capture using a membrane to separate CO_2 and H_2 in the gas stream of an IGCC power plant [48].

the reaction products (CO_2 and H_2) in the reactor vessel. Thus, to get high conversion efficiency of CO to CO_2, a catalyst is used and the WGS reaction is accomplished in two stages (and two vessels), with intermittent cooling of the syngas to help speed the reaction. This additional equipment and the associated energy requirements of the WGS process add to the cost of CO_2 capture.

One approach to reducing cost is the development of sorbents and membranes that can be used within a WGS reactor so that the shift reaction occurs with simultaneous capture of CO_2 [77]. Thus, in a sorbent-enhanced water-gas shift, the WGS catalyst is mixed with a CO_2 capture sorbent in a single reactor vessel. The sorbent removes CO_2 as soon as it is formed, which allows increased conversion of CO to CO_2. In this way, CO_2 capture is achieved simultaneously with an efficient WGS reaction, which can lower the overall capital cost of the system [78]. As with other sorbent-based capture schemes, however, the development of enhanced WGS reactors also requires a practical method of handling and regenerating the solid sorbent materials, which is a topic of ongoing research.

A similar concept for simultaneous WGS and CO_2 capture employs a membrane reactor in which either CO_2 or H_2 is separated as soon as it is formed [79]. Again, the removal of reaction products improves the effectiveness and speed of the WGS reaction. The possibility of using liquid solvents together with membranes also is being studied as a means of increasing the overall capture efficiency [48].

4.2.5. *Conceptual design stage*

At the conceptual design stage, most of the work related to pre-combustion capture is focused on improving the efficiency of the overall power plant, which in turn lowers the cost of CO_2 capture (see Section 2.5). Thus, improvements in all major IGCC system components—especially the air separation unit (ASU), gasifier and gas turbine— also are of interest for CO_2 capture. So too are studies of improved heat integration to reduce energy losses; advanced plant designs that integrate components such as the ASU and gas turbine air compressor; gasifier improvements that increase plant utilization; and advanced design concepts such as an IGCC system coupled with a solid oxide fuel cell. Examples of such studies can be found in several recent studies [12,80]. Fig. 22 shows an example of the cost reductions projected by the U.S. Department of Energy for conceptual designs of IGGC systems employing a variety of advanced technologies. These advances also would reduce the incremental cost of CO_2 capture. Substantial R&D efforts would be needed, however, to bring such designs to commercial reality.

4.2.6. *Conclusion for pre-combustion capture processes*

Many of research and development activities aimed at reducing the cost of pre-combustion CO_2 capture are similar in nature to those for post-combustion capture insofar as they involve the same basic concepts for new or improved gas separation processes. In addition, improvements are being sought in a variety of other IGCC plant components that also affect CO_2 capture costs, such as the air separation unit, gasifier, water-gas shift reactor and gas turbines. At the conceptual level, advanced plant designs employing new plant integration concepts and advanced technologies such as solid oxide fuel cells also are being actively investigated. The concepts that today appear most promising, however, are likely decades away from commercial reality.

4.3. *Status of oxy-combustion capture*

While commercial post- and pre-combustion CO_2 capture technologies are widely used in a variety of industrial applications to separate CO_2 from mixed gases, oxy-combustion as a CO_2 capture method is still under development and not yet used commercially for CO_2 capture. Here we briefly summarize the status of current oxy-combustion CO_2 capture technology developments.

4.3.1. *Commercial processes*

Although oxy-combustion carbon capture systems are not yet in commercial operation, the critical underlying technologies of oxygen production and oxygen combustion are mature and widely used in a variety of industrial settings. The largest industrial use of oxygen is for the smelting of iron ore in steel manufacturing, which consumes over half the commercially produced oxygen [81]. Here, high temperature oxy-combustion is used to oxidize excess carbon and other impurities in scrap iron or iron ore. The chemical

Table 15
Key advantages and challenges of membrane separation systems for pre-combustion CO_2 capture [48].

Membrane type	Description	Advantages	Challenges
H_2–CO_2 membranes	A membrane material which selectively allows either H_2 or CO_2 to permeate through the material; potential use in gasification processes with streams of concentrated H_2 and CO_2.	H_2 or CO_2 Permeable Membrane: No steam load or chemical losses. H_2 Permeable Membrane Only: Can deliver CO_2 at high pressure, greatly reducing compression costs. H_2 permeation can drive the CO shift reaction toward completion, potentially achieving the shift at lower-cost/higher temperatures.	Membrane separation of H_2 and CO_2 is more challenging than the difference in molecular weights implies. Due to decreasing partial pressure differentials, some H_2 will be lost with the CO_2. In H_2-selective membranes, H_2 compression is required and offsets the gains of delivering CO_2 at pressure. In CO_2 selective membranes, CO_2 is generated at low pressure, thus requiring added compression.
Membrane-Liquid Solvent Hybrids	Flue gas is contacted with a membrane and a solvent on the permeate side absorbs CO_2 and creates a partial pressure differential to draw CO_2 across the membrane.	The membrane shields the solvent from flue gas contaminants, reducing losses and allowing higher loading differentials between lean and rich solvent streams.	Capital cost associated with the membrane. Membranes may not keep out all unwanted contaminants. Does not address CO_2 compression costs.

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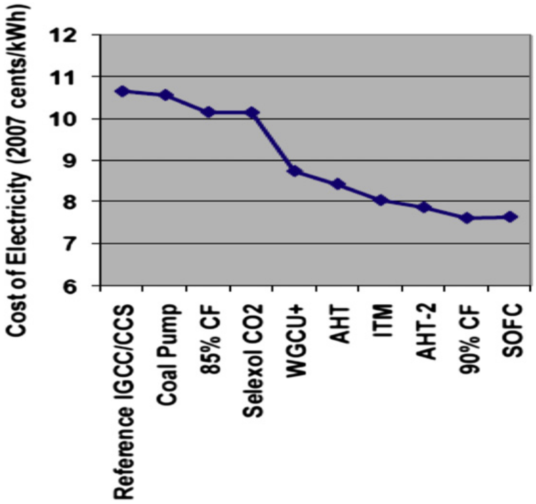

Fig. 22. Projected cost reductions for IGCC systems employing advanced technologies [80]. These improvements also reduce the cost of CO_2 capture. Terms not defined previously are: CF = capacity factor; WGCU = warm gas cleanup; AHT = advanced hydrogen-fired turbines (designs 1 and 2); ITM = ion transport membrane (for O_2 production); SOFC = solid oxide fuel cell (integrated with gasifier).

industry also uses substantial amounts of oxygen as an oxidizing agent to produce chemicals such as ethylene glycol. Perhaps more relevant to this paper, the combustion of oxygen or oxygen-enriched air in furnaces also is practiced commercially in some industries such as glass manufacturing. Thus, there is substantial industrial experience with oxygen and oxygen-enriched combustion systems. None of these industrial applications, however, separate or capture carbon dioxide from the resulting flue gas streams, so there is no direct industrial experience with oxy-combustion for large-scale CO_2 capture.

Most commercial air separation units (ASUs) employ a low-temperature cryogenic process to separate oxygen from other constituents of air (principally nitrogen and argon). The process can be scaled or deployed in multiple trains to deliver the quantities of oxygen required for a typical coal-fired power plant. A key drawback of current ASU technology, however, is its high energy requirements, which increase with the level of oxygen purity [82]. For a typical oxyfuel power plant design with 95 percent oxygen purity, Table 2 earlier showed that the energy penalty for oxygen production is similar to the penalty for amine solvent regeneration in post-combustion capture systems. Thus, for oxy-combustion carbon capture to be more economical, air separation methods are needed that are less energy-intensive than current cryogenic systems.

4.3.2. Full-scale demonstration plants

As with post- and pre-combustion capture, to date there have been no full-scale demonstrations of oxy-combustion CO_2 capture at a power plant, although several such demonstrations are planned. In the U.S., the Department of Energy announced in August 2010 a grant of $1 billion in federal cost-sharing for the FutureGen 2.0 project, under which an existing 200 MW oil-fired boiler at the Ameren power plant in Illinois will be replaced and repowered with a new supercritical coal-fired unit employing oxy-combustion and CO_2 capture. Outside the U.S., large-scale oxy-combustion demonstrations are planned in Canada, Germany, Spain and the U.K. (Table 16). These projects would employ a conventional ASU as the oxygen source, along with conventional flue gas cleanup systems where needed. Potentially, a flue gas desulfurization system may be omitted to reduce costs if it is determined that sulfur oxides can be safely co-sequestered with CO_2 without compromising either the boiler or pipeline operation.

Table 16
Planned large-scale demonstrations of oxy-combustion CO_2 capture [28–31].

Project name and location	Plant and fuel type	Planned year of startup	Plant size or capacity	Annual CO_2 captured (10^6 tonnes)
Projects in the U.S.				
FutureGen 2.0 (Meredosia, Illinois)	Coal-fired boiler	2015	200 MW	1.3
Projects outside the U.S.				
Boundary Dam (Estevan, Canada)	Coal-fired boiler	2015	100 MW	1.0
Datang Daqing (Heilongjiang, China)	Coal-fired boiler	2015	350 MW	~1.0
OXYCFB300 (Cubillos del Sil, Spain)	Coal-fired boiler	2015	300 MW	N/A
Oxy CCS Demonstration (North Yorkshire, UK)	Coal-fired boiler	2016	426 MW_g	~2.0

A key test for these demonstrations will be the integration of conventional ASUs to meet the oxygen needs of a large coal-fired boiler with substantial amounts of flue gas recirculation needed to control furnace temperatures under a variety of operating conditions. As noted earlier, alternative designs for new oxy-combustion plants have been proposed which would reduce or eliminate flue gas recirculation, such as by the use of a slagging combustor [83] or non-stoichiometric staged combustion [84], but to date there are no plans to demonstrate such approaches. Note too that all of the currently planned demonstration projects are around 100–200 MW in size, requiring only a single ASU train. Larger plants requiring more than 5000 tons per day of oxygen would need multiple ASUs, adding to the complexity and cost of the oxygen delivery system. As of this writing, it appears unlikely that all of the planned demonstrations will materialize in light of their cost and (in the case of the German plant) local public opposition to the project.

4.3.3. Pilot plant projects

Table 17 lists four European pilot plants currently testing oxy-combustion capture in an integrated system design at the pilot plant scale. Two plants now in operation each capture over 200 tons of CO_2 per day. Vattenfall's pilot plant at the Schwarze Pumpe power station in Germany (Fig. 23) is providing performance data needed to design the planned 250 MW demonstration plant listed in Table 16. The oxyfuel pilot plant operated by Total in France is of comparable size to the Vattenfall unit but operates on a gas-fired rather than coal-fired boiler. The most recent European project, in Spain, began operation in 2011 [85], but no results are publicly

Table 17
Pilot plant projects with oxy-combustion CO_2 capture [28–31].

Project name and location	Plant and fuel type	Year of startup	Plant size or capacity	Annual CO_2 captured (10^6 tonnes)
Schwarze Pumpe (Spremberg, Germany)	Coal-fired boiler	2008	30 MW_{th} (~10 MW)	0.075
Total Lacq (Lacq, France)	Natural gas-fired boiler	2009	30 MW_{th} (~10 MW)	0.075
OxyCoal UK (Renfrew, Scotland)	Coal-fired boiler	2009	40 MW_{th} (~13 MW)	N/A
CIUDEN (Cubillos del Sil, Spain)	Coal-fired boiler	2011	20 MW_{th} (~7 MW)	<0.092
CS Energy Callide A (Biloela, Australia)	Coal-fired boiler	2012	30 MW	0.3

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Fig. 23. Oxy-combustion pilot plant capturing CO_2 from the flue gas of a coal-fired boiler at the Schwarze Pumpe power station in Germany; Photo courtesy of Vattenfall.

available as of this writing. This project is intended to provide operational data for the large-scale OXYCFB300 power plant project listed in Table 16.

Not included in Table 17 are other pilot-scale facilities around the world that are also used to test various components of an oxy-combustion system, such as the 30 MW_{th} Clean Energy Development Facility of Babcock and Wilcox [86]. Similarly, Air Products is operating a pilot plant in Maryland that uses an experimental ion transport membrane (ITM) system for oxygen production, rather than a conventional cryogenic ASU [87]. That system, depicted in Fig. 24, is one of several new technologies under development that promises to deliver lower-cost oxygen.

While not a CO_2 capture technology, oxygen production is nonetheless the major cost and energy penalty item of an oxy-combustion system. For that reason, advanced methods of oxygen production are discussed in the following section on laboratory and bench-scale developments.

4.3.4. *Laboratory and bench-scale developments*

Laboratory and bench-scale R&D related to oxy-combustion is found worldwide and is focused mainly in the following areas [48]:

- Understanding oxy-combustion burner and boiler characteristics and their interactions with the overall system;
- Design of innovative oxy-combustion burners for new and retrofit applications;
- Development of improved flue gas purification technologies for oxy-fired systems;
- Development of lower-cost, high-efficiency oxygen production units; and
- Development of novel concepts such as chemical looping combustion.

Research topics include studies of fundamental mechanisms that affect the performance and design of oxygen-fired boiler systems,

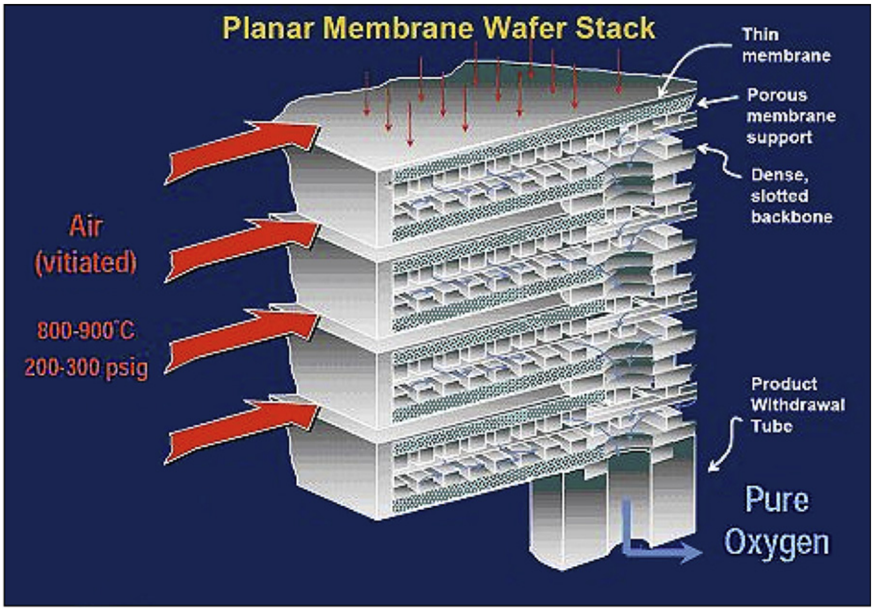

Fig. 24. The Ion Transport Membrane (ITM) oxygen production technology being developed by Air Products [88].

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such as oxy-combustion flame characteristics, burner design and fuel injection systems. Because of the high temperatures associated with oxygen combustion, development of advanced boiler materials is also a focus of research. In a number of areas, small-scale experiments are being coupled with computational fluid dynamic (CFD) modeling of oxy-combustion processes [48]. The development of advanced flue gas purification systems also is being pursued to find lower-cost ways to remove contaminants such as sulfur oxides, nitrogen oxides and trace elements such mercury. The ability to remove such pollutants during the CO_2 compression process is one of the potential innovations being studied [89].

There is a large body of literature that discusses and documents in detail the range of laboratory and bench-scale R&D activities and challenges in oxy-combustion CO_2 capture [90,91]. The remainder of this section elaborates briefly on two areas believed to offer the greatest promise for lower-cost capture.

4.3.4.1. Advanced oxygen production methods. Current commercial technology uses low-temperature (cryogenic) separation methods to produce high-purity oxygen. An alternative that promises a lower energy penalty and lower-cost is the ion transport membranes (ITM) system mentioned earlier. Here, thin non-porous membranes are used to separate oxygen from air at high temperature and pressure, as seen in Fig. 24. As with other membrane-based systems, the separation works on the principle of an oxygen pressure difference on either side of the membrane. The higher the pressure difference, the better is the separation. The goal is to produce ITM oxygen at one-third the cost and energy requirement of current cryogenic ASUs [88]. IGCC systems and other gasification-based processes are currently the most attractive applications for ITM oxygen since these processes already operate at the high pressures required by ITM technology. Oxy-combustion applications, however, would require the development of pressurized combustion systems in order to take full advantage of ITM oxygen production.

Unlike ITMs, which separate oxygen based on a pressure differential, the oxygen transport membrane (OTM) concept utilizes the chemical potential of oxygen as the driving force. The advantage of this approach is that it can be integrated directly into a boiler, with air on one side of the membrane and fuel combustion on the other side. Combustion decreases the oxygen concentration, which increases the chemical potential difference to drive O_2 through the membrane. This process is still in the early stages of development [48]. A review of recent research in this area can be found in Hashim et al. [92].

Other new oxygen production methods being investigated use solid sorbents to absorb oxygen from air. The sorbent material is then transferred to another vessel where it is heated, releasing the oxygen. This is fundamentally the same approach discussed in Sections 4.1 and 4.2 for CO_2 capture using solid sorbents. For oxygen production the sorbent material and process conditions are different. The process called ceramic autothermal recovery uses the mineral perovskite. It releases heat while adsorbing oxygen from air. This heat potentially could be used along with heat from power plant flue gases to reducing the overall energy penalty of oxygen production. Another sorbent being investigated is manganese oxide, which absorbs oxygen from high pressure air passed over the sorbent. This technology is potentially easier to build and lower in cost [48]. Until a larger-scale process is developed and tested, however, cost estimates remain highly uncertain.

4.3.4.2. Chemical looping combustion. Another novel oxy-combustion technology being developed is chemical looping combustion (CLC). This is similar to the sorbent-based oxygen production method discussed above. Here, however, the oxygen-carrying sorbent—typically a metal oxide—is contacted with a fuel, so that combustion occurs. The resulting exhaust stream contains only carbon dioxide and water vapor, as in other oxy-combustion schemes. A schematic of this concept is shown in Fig. 25.

Chemical looping has the potential to make carbon capture significantly cheaper than current systems. However, it is still at an early stage of development, with challenges in materials handling and oxygen carrier selection. Currently the largest chemical looping combustor is a 120-kW unit being tested in Austria [46]. Projects funded by the U.S. Department of Energy include two chemical looping tests, one by Alstom using calcium compounds as an oxygen carrier, the other by Ohio State University using an iron oxide carrier [93]. Alstom is currently testing prototypes of 1 MW_{th} and 3 MW_{th} in Germany and US, respectively [94].

4.3.5. *Conceptual design stage*

As with pre-combustion CO_2 capture systems, a substantial amount of current activity on oxy-combustion capture is still at the conceptual design stage, positing and analyzing alternative system configurations to maximize overall efficiency and minimize estimated cost. Conceptual designs encompass a broad range of fuels and power systems. Many of these designs include advanced component technologies and heat integration schemes that do not currently exist, but which illustrate the potential for process improvements.

For example, Anantharaman et al. [95] propose a novel oxy-combustion cycle for natural gas-fired power plants that combines an oxygen transport membrane with advanced heat integration in a reactor design (Fig. 26) that theoretically achieves 85 to 100 percent CO_2 capture with a higher plant efficiency than a current NGCC plant with CO_2 capture. Other oxy-combustion designs for combined cycle power plants utilize CO_2 instead of air to generate power from advanced gas turbines, or employ ITM technology to achieve high-efficiency power generation with high CO_2 capture [7,96]. All of these advanced concepts, however, require the (costly) development and integration of advanced technologies that do not yet exist and which may have only limited market potential. Thus, despite their theoretical advantages, it appears unlikely that such concepts will advance to the later stages of technology development any time soon.

Other conceptual designs for coal-fired power plants seek improved methods of heat and process integration to improve overall plant efficiency using conventional technology for power

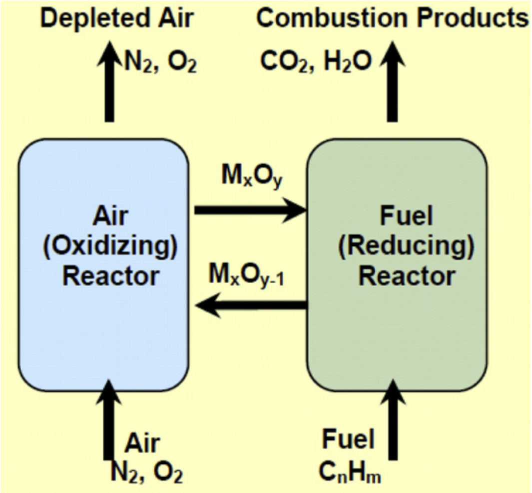

Fig. 25. Schematic of a chemical looping combustion (CLC) process [48].

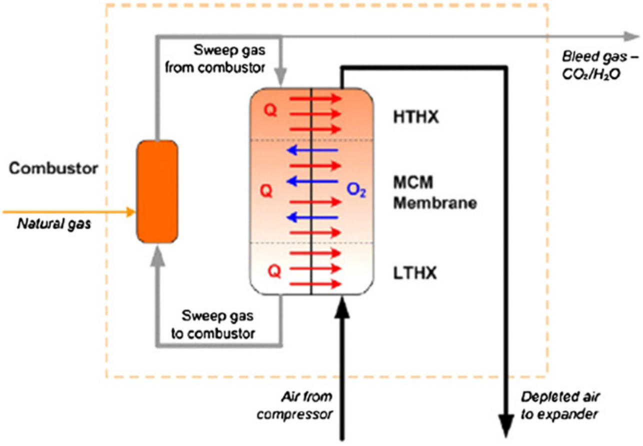

Fig. 26. A proposed oxygen-mixed conduction membrane reactor design for a natural gas-fired power plant [95].

generation and oxygen production [90,97]. More advanced concepts envision pressured combustion with oxygen as a preferred approach for achieving high-efficiency along with lower-cost CO_2 capture. These analyses based on thermodynamics and optimization methods are useful for identifying the most promising concepts to consider for further development.

4.3.6. *Conclusion for oxy-combustion processes*

Arguably the most compelling need—and a major focus of R&D for oxy-combustion systems —is for improved, lower-cost processes to deliver large quantities of oxygen, the major cost item in current oxyfuel schemes. Some of the promising approaches are similar to those discussed for post- and pre-combustion capture, such as the use of membrane separation processes. The development of improved gas cleaning processes to remove traditional air pollutants is also a focus of current R&D. To the extent that oxy-combustion systems are able to transport and sequester multi-pollutant gas streams including SO_2, NO_x and other trace species found in power plant flue gas streams, costs could be further reduced by avoiding the need to remove such pollutants. At the conceptual level, advanced plant designs employing new plant integration concepts and advanced technologies such as chemical looping combustion also are being actively investigated and appear promising. Because they are at the early stages of development, however, it remains to be seen which if any of these concepts eventually develops into a viable commercial technology.

5. Cost and deployment outlook for advanced capture systems

Having reviewed a number of advanced technology developments, we now address two key questions: (1) how much performance improvement and cost reduction is expected from the CO_2 capture technologies now under development? (2) When will these advanced technologies be available for commercial use? To address the first question we first examine results from recent studies by DOE and others of projected cost reductions for power plants with advanced capture systems. To address the second question, Section 6 show a set of technology roadmaps and deployment scenarios developed by governmental and private organizations involved in CO_2 capture technology R&D. Then, to gain perspective on these projections, Section 7 reviews the experience of past R&D programs to develop advanced capture technologies for power plant emissions.

5.1. *Projected cost reductions for CO_2 capture*

Fig. 9 earlier summarized the range of cost estimates for power plants using current technology for CO_2 capture and storage. Other sources discuss in detail the many factors that affect such estimates [7,98]. In the context of the present paper, it is especially important to emphasize the uncertainty inherent in any cost estimate for a technology that has not yet been built, operated and replicated at a commercial scale. In general, the further away a technology is from commercial reality, the cheaper it tends to look. This is illustrated graphically in Fig. 27, which depicts the typical trend in cost estimates for a technology as it advances from concept to commercial deployment.

Keeping in mind this uncertainty, we summarize the results of several recent studies that estimated potential cost reductions from technology innovations both in CO_2 capture processes as well as in other power plant components that influence CO_2 capture cost. These studies employ two conceptually different methods of estimating future costs. The “bottom up” method uses engineering analysis and costing to estimate the total cost of a specified advanced power plant design. In contrast, the “top down” method uses learning curves derived from past experience with similar technologies to estimate the future cost of a new technology based on its projected installed capacity at some future time. The latter parameter represents the combined effect of all factors that influence historically observed cost reductions (including R&D expenditures, learning-by-doing and learning-by-using).

5.2. *Results from engineering-economic analyses*

Fig. 28 shows the results of a 2006 analysis by DOE of potential advances in the major CO_2 capture routes [100]. Results are shown for PC plants and IGCC plants. The bars in Fig. 28 show the percent increase in the total cost of electricity (COE) compared to the same plant type without CO_2 capture. As more advanced technologies are implemented the incremental cost is reduced significantly. On an absolute basis, the total cost of electricity generation falls by 19 percent for the IGCC cases and by 28 percent for the PC cases. The biggest cost reductions come in the final steps for each plant type. However, the technologies in those cases are still in the early stages of development, including advanced solid sorbents for CO_2 capture, membrane systems for water-gas shift reactors and chemical looping for oxygen transport. As suggested earlier in Fig. 27, cost estimates for these cases are the least reliable and most likely to escalate as the technology approaches commercialization.

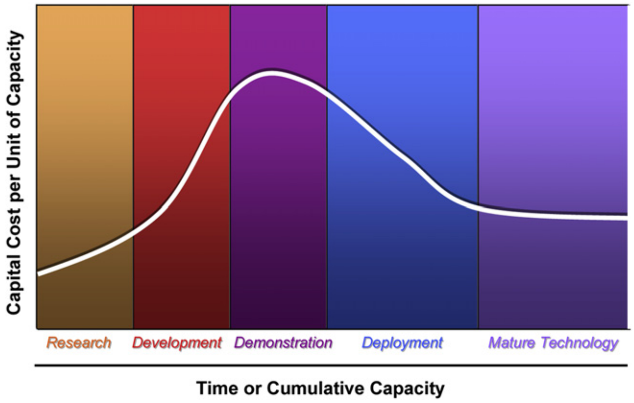

Fig. 27. Typical cost trend for a new technology as it develops from a research concept to commercial maturity Adapted from [99].

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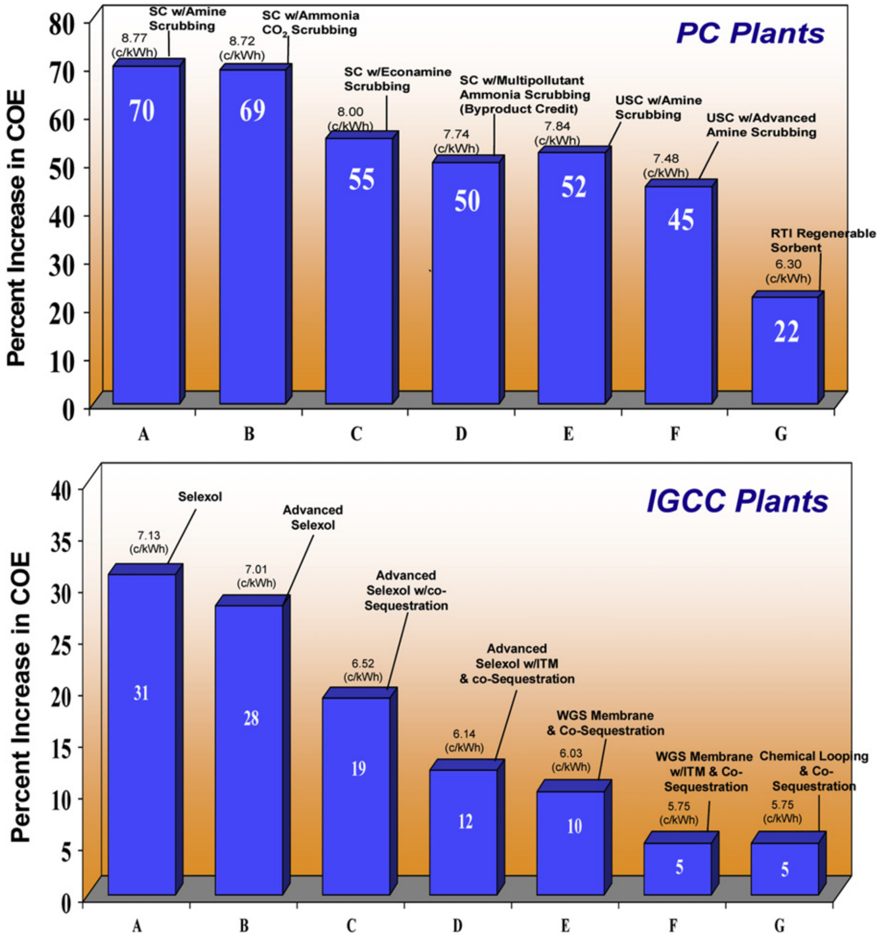

Fig. 28. Cost of electricity (COE) increases for power plants with CO_2 capture and storage using current technology (column A) and various advanced technologies (column B to G). The value of total COE appears at the top of each column. (Abbreviations: SC = supercritical; USC = ultrasupercritical; RTI = Research Triangle Institute; ITM = ion transport membrane; WGS = water-gas shift) [100].

The 2006 DOE analysis also included four oxy-combustion cases (not shown in Fig. 28) in which the COE for an advanced system fell by 19 percent (from a 50 percent increase in COE for a current supercritical PC plant, to a 21 percent increase for advanced SCPC with ITM oxygen production). Because oxy-combustion systems are still under development and not yet demonstrated at a commercial scale, assumed plant configurations and cost estimates for these systems are more uncertain and variable than for current pre- and post-combustion systems. For example, while some studies show oxy-combustion for new power plants to be somewhat lower in cost than post-combustion capture [101] others report it to be higher in cost [7]. There is general agreement, however, that continued R&D can reduce the future cost of all these systems.

Fig. 29 shows a more recent (2010) DOE analysis of potential reductions in capture cost from sustained R&D [102]. Here, the total cost of a new supercritical PC plant with CCS declines by 27 percent

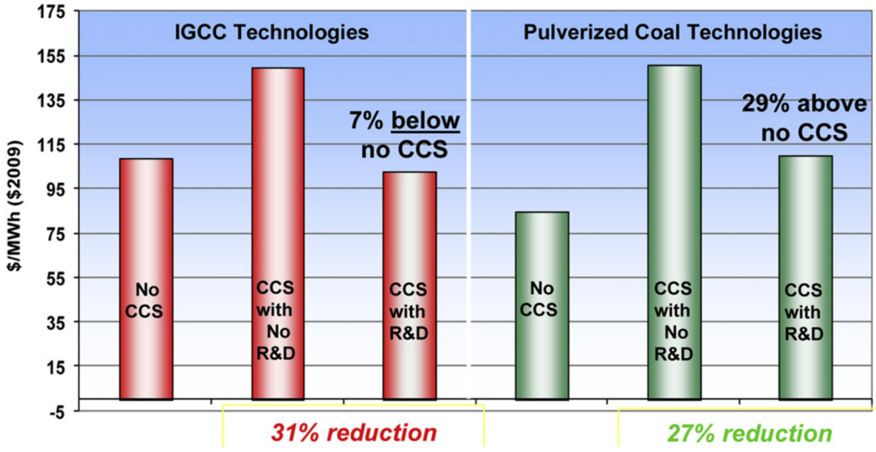

Fig. 29. Current cost of electricity (COE) for IGCC and PC power plants with and without CO_2 capture and storage, plus future costs with advanced technologies from R&D [102].

while the IGCC plant cost falls by 31 percent. As a result, the future IGCC plant with CCS costs 7 percent less than the current IGCC plant without capture. For the PC plant the CCS cost penalty falls by about half in this analysis.

Since many of the components assumed in the DOE analysis are still at early stages of development, cost estimates for these advanced technologies are again highly uncertain. Nonetheless, these estimates can be taken as a rough (perhaps optimistic) indication of the potential cost savings that might be realized. Other organizations have estimated similar cost reductions for advanced plant designs with CCS (e.g., see summary in [7]).

Typically missing from engineering-based cost estimates such as these is an indication of the time frame in which these advanced technologies are expected to be in commercial use. This is especially problematic for environmental technologies like CO_2 capture processes since the market for such systems depends mainly on government policies that require or incent their use. An alternative approach to forecasting technology costs, based on learning or experience curves, comes closer to providing a temporal dimension together with cost estimates, as discussed below.

5.3. *Results from experience curve analyses*

This approach models the future cost of power plants with CCS as a function of the total installed capacity of such plants. While time is not an explicit variable, it is implied by the choice of total plant capacity that is projected. The future cost reductions shown in Fig. 30 are from a detailed analysis that applied historical learning rates for selected technologies to the components of four types of power plants with CO_2 capture (PC, NGCC, IGCC and oxyfuel) [82]. The component costs were then summed to estimate the future cost of the overall power plant as a function of new plant capacity. The analysis also considered uncertainties in key parameters, including potential increases in cost during early commercialization.

Fig. 30 shows the resulting ranges of cost reduction estimated for each of the four types of power plants with CO_2 capture after an assumed deployment of 100,000 MW for each system worldwide (roughly equal the worldwide capacity of FGD systems two decades after that technology was first introduced at U.S. power plants). These results reflect different levels of maturity for each plant type as well as the CO_2 capture system. Thus, the IGCC plant—whose principal cost components are less mature than those of combustion-based plants—shows the largest potential for overall cost reductions. The combustion-based plants show a smaller potential since most of their components are already mature and widely deployed. In all cases, however, the incremental cost of CO_2 capture system falls more rapidly than the cost of the overall plant. The high end of the cost reduction ranges in Fig. 30 is similar to DOE's "bottom up" estimates shown earlier. The low end of the ranges, however, is smaller by factors of 2–3. That result suggests a more gradual rate of cost reductions from continual improvements to capture technologies as CCS is more widely deployed.

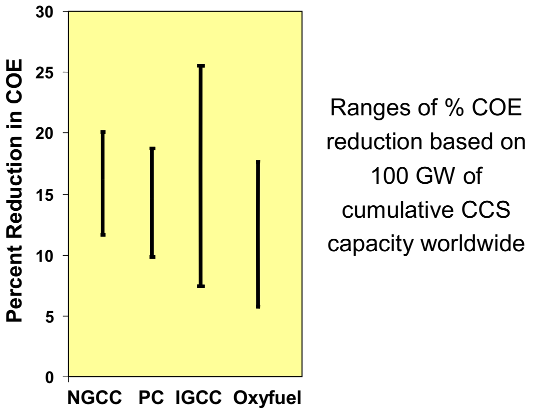

Fig. 30. Projected cost reductions for four types of power plants with CO_2 capture based on experience curves for major plant components [82].

6. Roadmaps for capture technology commercialization

Here we look at projected timetables for the development and commercialization of CO_2 capture systems. Such "roadmaps" have been developed by a number of governmental and private organizations involved in CO_2 capture technology R&D. They provide a current perspective on the time frame in which various organizations anticipate that improved or lower-cost capture systems will be commercial and available for use at power plants and other industrial facilities. None of the organizations surveyed, however, make explicit assumptions about the nature and timing of government policies that require or incentivize the use of CCS to abate carbon emissions. Absent such policy drivers, the commercial deployments projected in some of the roadmaps below are unlikely to materialize, as elaborated later in Section 7.

6.1. *The DOE roadmap*

As part of its Carbon Sequestration Program, the U.S. Department of Energy (DOE) has developed and periodically updates a roadmap displaying the projected timetable for major program elements, including CO_2 capture technology development [103]. Fig. 31 shows an excerpt from the most recent DOE roadmap published in 2010 [104]. Fig. 32 shows a more detailed timeline for advanced CO_2 capture technologies applied to existing plants.

The 2010 DOE roadmap has a timeline extending to 2030, roughly a decade longer than the previous (2007) roadmap. The more detailed roadmap for advanced post-combustion capture systems in Fig. 32 anticipates commercial deployment of some advanced technologies in 2020, with large-scale demonstrations beginning four years earlier [104]. Laboratory and bench-scale R&D would, on average, advance to pilot-scale testing after about two years, with subsequent pilot plant testing and scale-up prior to large-scale demonstrations.

6.2. *The EPRI and CURC roadmap*

The Electric Power Research Institute (EPRI) carries out R&D on behalf of member utility companies. EPRI-supported projects include development and testing of advanced carbon capture technologies. Fig. 33 shows a roadmap developed jointly between EPRI and the Coal Utilization Research Council (CURC), an industry advocacy group that promotes the efficient and environmentally-sound use of coal. Recent updates to this roadmap call for four demonstrations of IGCC with CCS by 2025, plus nine demonstrations of combustion with CCS by 2025 [105]. Like the DOE plan, the CURC-EPRI roadmap expects CO_2 capture systems for power plants to be commercial by 2020. That roadmap, however, shows a heavier reliance on continued improvements to technologies that are already at the advanced stages of development.

EPRI researchers also have put forth a timeline for carbon capture developments based on the Technology Readiness Levels (TRLs) described earlier in Section 3.2. This timeline, shown in Fig. 34, characterizes most systems being developed today at TRLs 5 through 7. It shows activity at TRL 8 (equivalent to large-scale demonstration projects) beginning in 2010, with commercial scale plants (TRL 9) coming online by 2018. This implies a 10–15 year development schedule from concept to

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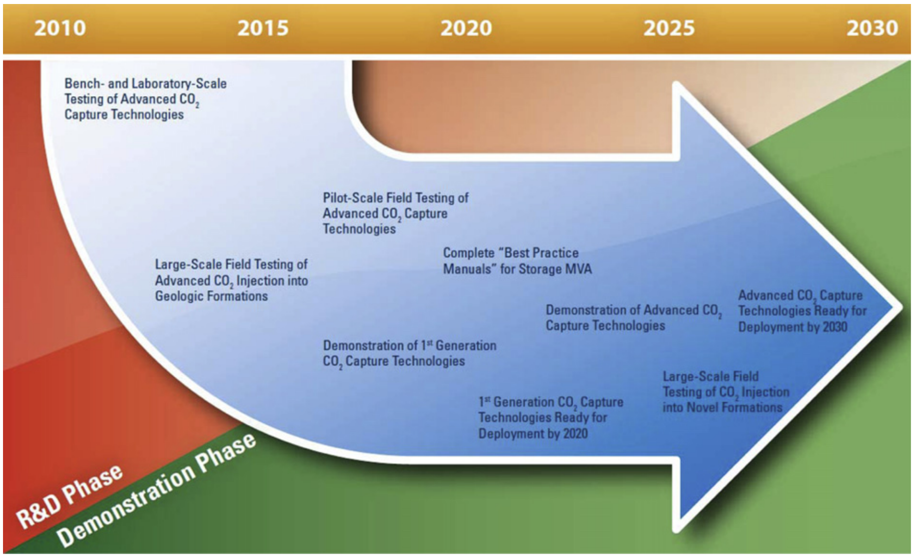

Fig. 31. The DOE carbon capture and storage RD&D roadmap [104].

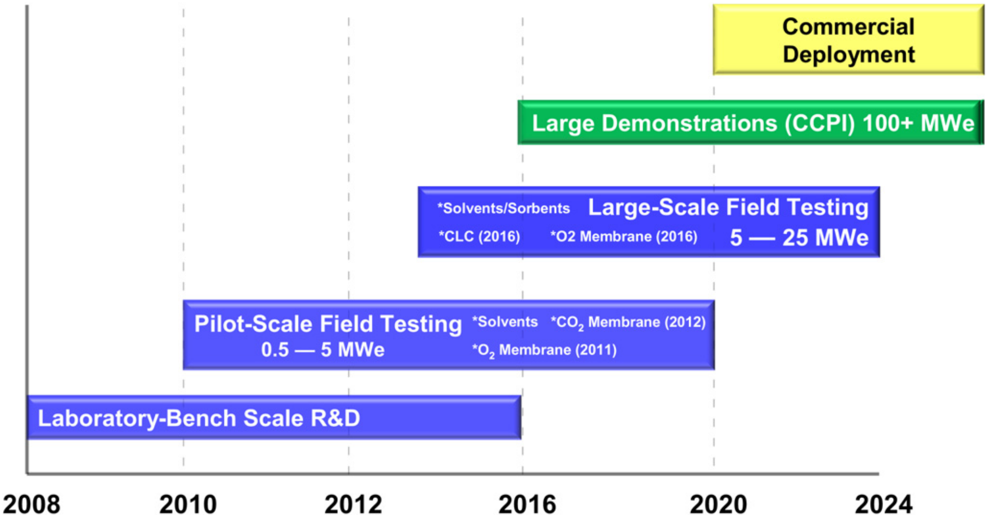

Fig. 32. DOE's timeline from R&D to commercial deployment of advanced post-combustion capture technologies for existing power plants [103].

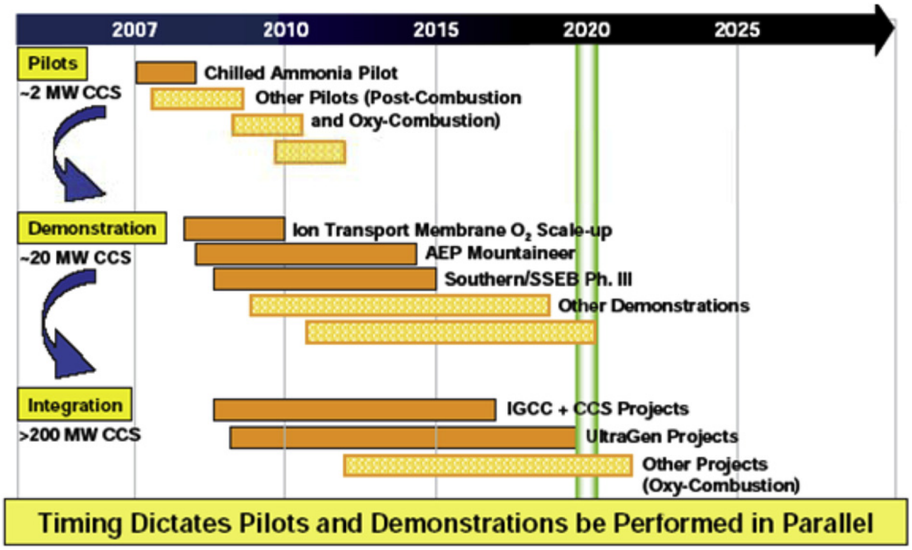

Fig. 33. Steps in technology validation and scale-up projects to meet CURC-EPRI roadmap goals for advanced coal technologies with CCS [105].

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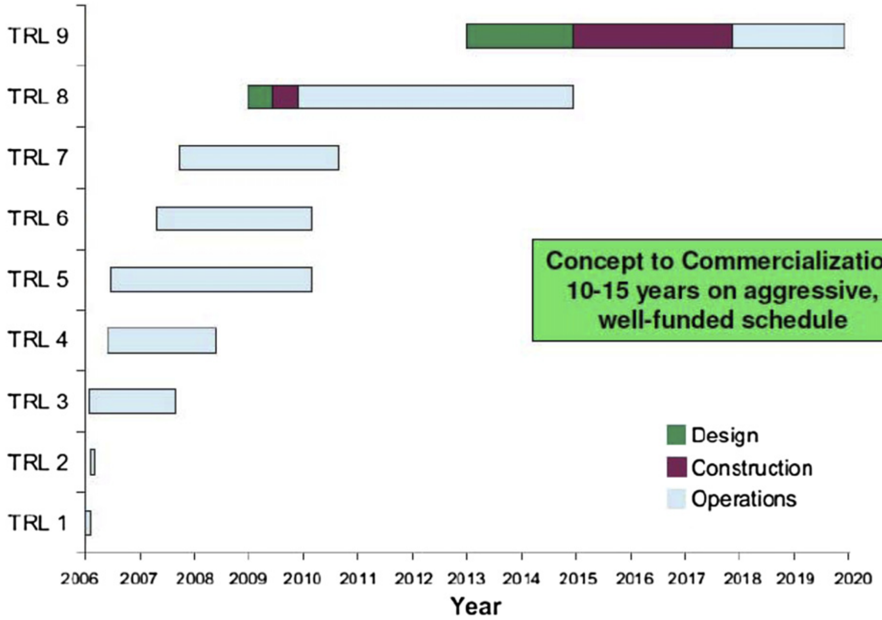

Fig. 34. EPRI projections of capture technology development based on technology readiness levels (TRLs) [47].

commercialization. EPRI acknowledges that this schedule represents an aggressive and well-funded program of RD&D. It does not comment on whether current RD&D programs meet those criteria.

6.3. *The CSLF roadmap*

The Carbon Sequestration Leadership Forum (CSLF) is an international initiative that is focused on the development of improved cost-effective technologies for CO_2 capture and storage. Its mission is to facilitate the development and deployment of such technologies via collaborative efforts [106]. The CSLF roadmap in Fig. 35 sets out development goals in three time periods: 2009–2013, 2014–2020 and 2020 and beyond. For CO_2 capture the goal for the first stage is "development of low-cost and scalable carbon capture technologies." Goals for the second stage involve full-scale demonstrations of these technologies, while the goal for 2020 and beyond is to have these technologies deployed commercially. The roadmap also lays out goals for CO_2 transport and storage as well as the development of integrated full-scale CCS projects by 2013. As an international organization the CSLF does not itself provide funding for CO_2 capture R and D; rather, it relies on country-level support for such projects.

6.4. *Other roadmaps and milestones*

Several other international groups and organizations have set goals and targets for the demonstration, commercialization and deployment of CO_2 capture and storage systems. At its 2008 summit meeting, the Group of Eight (G8) heads of state—representing the governments of Canada, France, Germany, Italy, Japan, Russia, the United Kingdom and the United States—committed to "strongly support the launching of 20 large-scale CCS demonstration projects globally by 2010, … with a view to beginning broad deployment of CCS by 2020" [107]. This action was based on recommendations of the CSLF and the International Energy Agency (IEA).

In conjunction with its global energy modeling activities, the IEA also has published a CCS Roadmap calling for increasing numbers of pilot and demonstration plants worldwide through 2035 [108]. To support the commercialization of CCS globally, the IEA sees a requirement for about 30 such new-build pilot and demonstration projects in the 2020–2025 time frame, an additional 100 projects in 2025–2030 and about 40 more in 2030–2035. A majority of early large-scale projects would take place in countries of the OECD (Organization for Economic Cooperation and Development), but after 2030 non-OECD countries would take the lead in commercializing CCS plants.

6.5. *Scenarios for CCS deployment*

Research groups worldwide have developed and used a variety of energy-economic models to predict the consequences of national and international policies to mitigate global climate change (e.g., [4,109]). These studies typically assume that CCS is available for deployment at power plants and other industrial facilities by at least 2020. Fig. 36 shows recent results from five different models used to project the least-cost U.S. energy mix in 2050 in response to

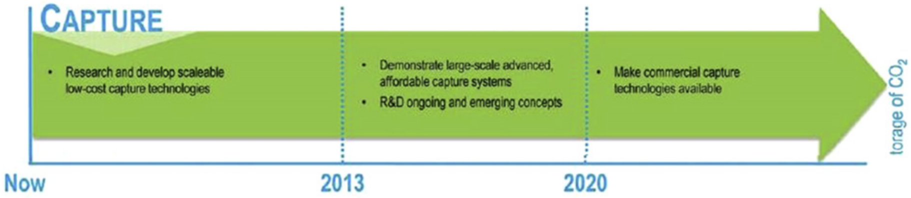

Fig. 35. Key milestones in the CSLF technology roadmap [106].

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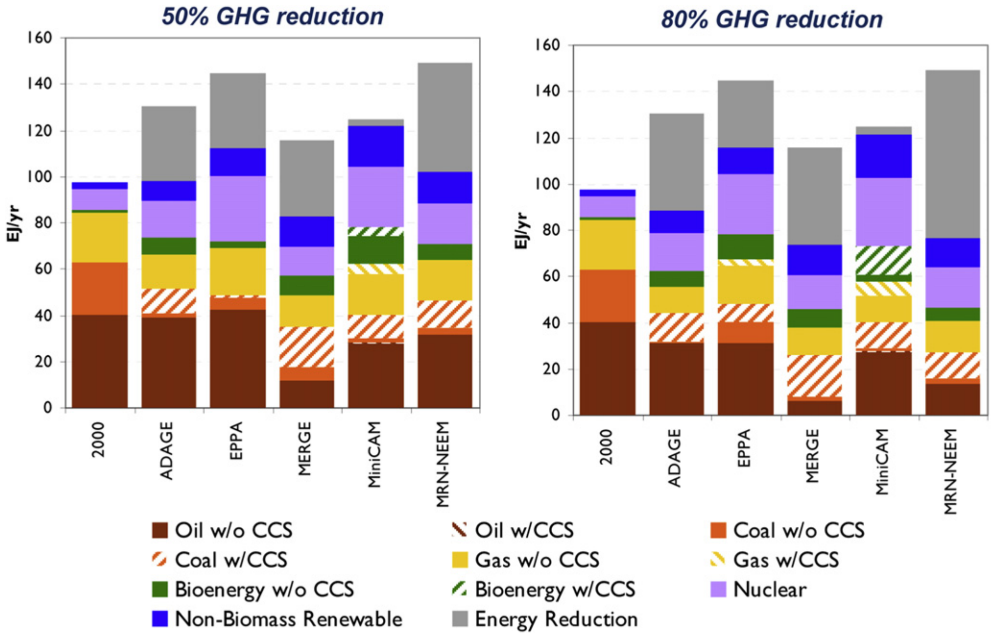

Fig. 36. Actual 2000 energy use and projected least-cost U.S. energy mix in 2050 for two scenarios of GHG reductions (below 1990 levels). The cross-hatched areas indicate facilities with CCS [109].

policy scenarios requiring national reductions in greenhouse gas emission of 50 percent to 80 percent below 1990 levels [109]. Results for the five models show different levels of deployment of CCS relative to other GHG reduction options in each scenario, illustrating the uncertainty in future demand for CO_2 capture technology. All models, however, deploy CCS more extensively as the emission reduction requirement grows more stringent—not only at coal-fired power plants but at natural gas-fired and biomass-fired plants as well. This indicates the potential importance of CCS as a cost-effective mitigation option for achieving climate goals. Without a strong policy driver, however, there is little or no demand for this technology.

6.6. *Conclusion for CCS roadmaps*

Current roadmaps and scenarios for carbon capture technology commercialization and deployment envision that improved, lower-cost capture systems will be generally available for use at power plants and other industrial facilities by 2020. At the same time, both public and private-sector research organizations acknowledge that a sustained R&D effort will be required over the next decade to achieve that goal—especially for many of the promising new processes that are still in the early stages of development. The magnitude of future cost reductions also is likely to depend on the pace of CCS technology deployment as well as on continued R&D support. The next section of this paper looks at past experience with other power plant environmental technologies to provide additional perspectives on the pace of new technology development, deployment, performance improvements and cost reductions.

7. Lessons from past experience

In this section we look retrospectively at a number of other recent efforts to develop and commercialize advanced technologies to improve the effectiveness and lower the cost of air pollutant capture at coal-fired power plants. The purpose of this analysis is to glean insights that are useful for assessing realistically the prospects for improved, lower-cost CO_2 capture systems. First we present several case studies of prior DOE supported efforts to develop novel, lower-cost systems to capture power plant sulfur dioxide (SO_2) and nitrogen oxide (NO_x) emissions. These past efforts bear a number of similarities to current efforts for CO_2 capture systems. Thus, they provide some historical benchmarks on the time required to bring a new process from concept to commercialization and the factors that influence the probability of success.

Following this we present some historical data on the rates of technology deployment, performance improvements and cost reductions for post-combustion capture system of SO_2 and NO_x. Again, the purpose is to provide benchmarks for assessing current projections for CO_2 capture systems. The critical role of government policies in establishing markets for environmental technologies also is discussed and illustrated with examples drawn from past experience with post-combustion SO_2 and NO_x capture technologies.

7.1. *Case studies of novel capture technology development*

Current efforts to develop new or improved carbon capture systems are in many respects similar to efforts that began in the late 1970s to develop improved, lower-cost technologies for power plant SO_2 and NO_x controls. Those activities followed passage of the 1970 Clean Air Act Amendments (CAAA) and the adoption of federal New Source Performance Standards (NSPS) requiring “best available control technology” for major new sources of air pollution, including fossil fuel power plants. Although SO_2 capture technology had been used commercially since the early twentieth century on various industrial processes (such as metal smelters), it had seldom been used to desulfurize power plant flue gases, and only then outside the U.S. The same was true of post-combustion NO_x capture technologies.

By the late 1970s, in response to NSPS and CAAA requirements, the most widely used technology for post-combustion SO_2 control was a flue gas desulfurization (FGD) system or “scrubber” that used a slurry of water and limestone to capture SO_2 via chemical reactions. These early “wet FGD” systems had high capture efficiencies (up to about 90 percent), but were widely regarded as being very expensive, difficult to operate reliably and with a high energy

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penalty [110]. In the case of nitrogen oxides, post-combustion capture systems such as selective catalytic reduction (SCR) were deemed in the 1970s to be too costly and unavailable to be required under the NSPS. Instead a less stringent requirement was imposed that did not require post-combustion capture, but rather could be met using only low-NO_x burners [111].

By the 1980s, U.S. coal-fired power plants were being targeted for further reductions in SO_2 and NO_x emissions to curtail the growing problem of acid deposition (acid rain). In response, DOE launched major initiatives to develop "high risk, high payoff" technologies that promised significant cost reductions in power plant SO_2 and NO_x emissions control compared to the prevailing FGD and SCR technologies.

Five new technologies supported under the DOE Clean Coal Technology program are briefly described below. Three of the novel processes involved post-combustion SO_2 and NO_x capture in a single process rather than in separate units. The other two processes sought more cost-effective SO_2 capture by injecting solid sorbents directly into the power plant furnace or flue gas duct. Of particular relevance to this paper are the time required to develop each process and its ultimate fate in the commercial marketplace.

7.1.1. The copper oxide process

The use of copper oxide as a sorbent for sulfur removal was first investigated at the laboratory-scale by the U.S. Bureau of Mines in 1961 [112]. Pilot-scale tests were performed in the mid-1960s and by 1973 the process saw industrial use for sulfur removal at a refinery in Japan [39]. DOE continued to develop the process as a lower-cost way to remove both SO_2 and NO_x from power plant flue gases, while producing sulfur or sulfuric acid as a byproduct in lieu of solid waste [113]. Fig. 37 shows several milestones in the process development.

After a series of design changes following pilot plant tests in the 1970s and 1980s, DOE began developing designs for a 500 MW power plant in the 1990s and planned a new 10 MW pilot plant as part of its Low Emission Boiler System project [114]. However, by the time the required Environmental Impact Statement for that project was completed, the copper oxide process had been replaced by a conventional wet FGD system [115]. Although the process never developed into a commercial technology for combined SO_2 and NO_x capture, research on copper oxide sorbents still continues [116,117].

7.1.2. The electron beam process

The electron beam process for flue gas treatment was first introduced by the Ebara Corporation of Japan in 1970 [39]. The concept was that energy from the electron beam would excite chemicals in the flue gas, causing them to break down and form other stable compounds. The process was promoted as a more cost-effective way to simultaneously capture both SO_2 and NO_x with high (~90 percent) efficiency.

By 1977, Ebara's testing moved to the pilot plant scale and in 1985 their subsidiaries in the U.S. and Germany opened two more pilot plants, one in Indiana and one in Germany [118]. DOE provided partial funding for the U.S. facility. Continued R&D led to the first commercial plant in China in 1998, nearly three decades after the process was first envisioned. This was followed by three more plants built between 1999 and 2005, one in Poland, the other two in China [119]. The overall cost of this system is highly dependent on the market value of the ammonium sulfate and ammonium nitrate by-products that are produced, as well as on the cost of ammonia, the key reagent for the process. The need for these byproduct chemicals may help explain the adoption of this process in China. However, there have been no commercial installations of the electron beam process in the United States.

7.1.3. The NOXSO process

The NOXSO process was another concept for post-combustion capture of both SO_2 and NO_x from power plant flue gases. It used a solid sorbent of sodium carbonate supported on alumina beads. The sorbent chemistry was based on an alkalized alumina process developed by the U.S. Bureau of Mines in the 1960s. A novel feature

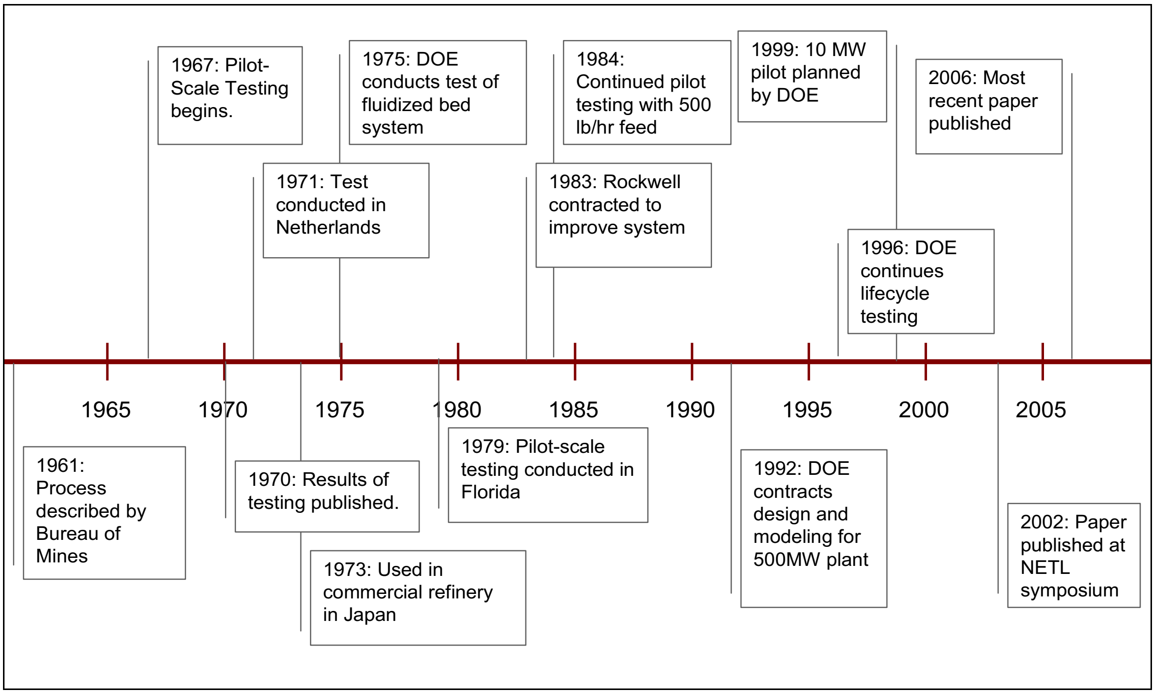

Fig. 37. Development history of the copper oxide process for post-combustion SO_2 and NO_x capture.

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the NOXSO process was the use of a fluidized bed reactor for sorbent regeneration. Fig. 38 shows the process development timeline, which began in 1979 with funding from DOE.

Pilot plant and life cycle testing were carried out from 1982 to 1993. In 1991 the newly-created NOXSO Corporation received a DOE contract to build a commercial scale demonstration plant [120]. However, a number of administrative problems ensued, leading to several changes in the project site location. A legal dispute with the owner of the final project site culminated in the bankruptcy and subsequent liquidation of the NOXSO Corporation [121]. After two decades of development the process was never demonstrated at scale or subsequently commercialized.

7.1.4. *The furnace limestone injection process*

In the early 1980s, the prospect of new restrictions on SO_2 emissions to control acid rain prompted interest in sulfur removal methods that were more cost-effective than commercial post-combustion capture systems, especially for existing power plants. The furnace limestone injection process promised to be such a technology. Limestone sorbent would be injected directly into the furnace and react with sulfur oxides to achieve moderate (roughly 50 percent) removal efficiencies, but at very low-cost. The method was first tested by Wisconsin Power in 1967 [122]. In the 1980s and 1990s, DOE supported two methods of furnace sorbent injection (called LIFAC and LIMB), as seen in Fig. 39.

The LIFAC (Limestone Injection into the Furnace and Activation of Calcium oxide) process combined limestone injection with a humidification system to capture SO_2 [123]. First developed by the Tampella Company in 1983, it was later tested at a commercial scale in Finland. DOE supported demonstrations in the U.S. starting in 1990, achieving 70 to 80 percent sulfur removal rates. The LIMB (limestone injection with multi-stage burners) process was first developed by the U.S. Environmental Protection Agency. It achieved approximately 50 percent SO_2 removal using limestone, with somewhat higher capture efficiencies using more expensive lime sorbents. Testing of both processes encountered failures of the electrostatic precipitator at the test plants due to the larger volume of solids being collected. Technical solutions added to the cost [124,125]. The LIFAC process was eventually demonstrated and used commercially at nine facilities outside the U.S. as indicated in Fig. 39. However, neither LIFAC nor LIMB was adopted commercially for SO_2 control in the United States. Other SO_2 reduction options (mainly switching to low-sulfur coals) proved to be more cost-effective for achieving moderate reductions in SO_2 emissions.

7.1.5. *The duct sorbent injection process*

Duct sorbent injection (DSI) is another post-combustion SO_2 capture concept similar to furnace limestone injection, except that the sorbent is injected into the flue gas duct after the boiler where temperatures are lower and physical access is generally easier. This was proposed as a simpler and more cost-effective method of achieving modest SO_2 reductions at existing power plants. Fig. 40 shows the process development timeline.

Babcock and Wilcox began work on a DSI system in 1980 for their SOx-NO_x-ROx-Box (SNRB)—a combined pollutant control system which DOE tested twelve years later. Pilot and demonstration projects of DSI for SO_2 capture during the 1980s and early 1990s achieved capture rates rarely exceeding 40 percent with calcium-based sorbents. Costs and technical complexity were similar to the more effective furnace injection systems [126]. Subsequent process modifications improved the SO_2 capture efficiency, but at a higher cost. There were no commercial adoptions of DSI following the DOE test programs.

In 1996, DSI was retooled for use in mercury control. It developed into the TOXECON process, which was tested at full-scale in 2003, achieving 90 percent capture of flue gas mercury [127]. Duct sorbent injection for mercury control is now offered commercially but has not been widely adopted in light of continuing uncertainty over U.S. power plant mercury emissions regulations.

7.2. *Implications for advanced carbon capture systems*

Several lessons can be gleaned from the case studies above that are relevant to current efforts to develop lower-cost carbon capture systems for power plants. The first is the importance of markets for new environmental technologies. Just as with advanced CO_2

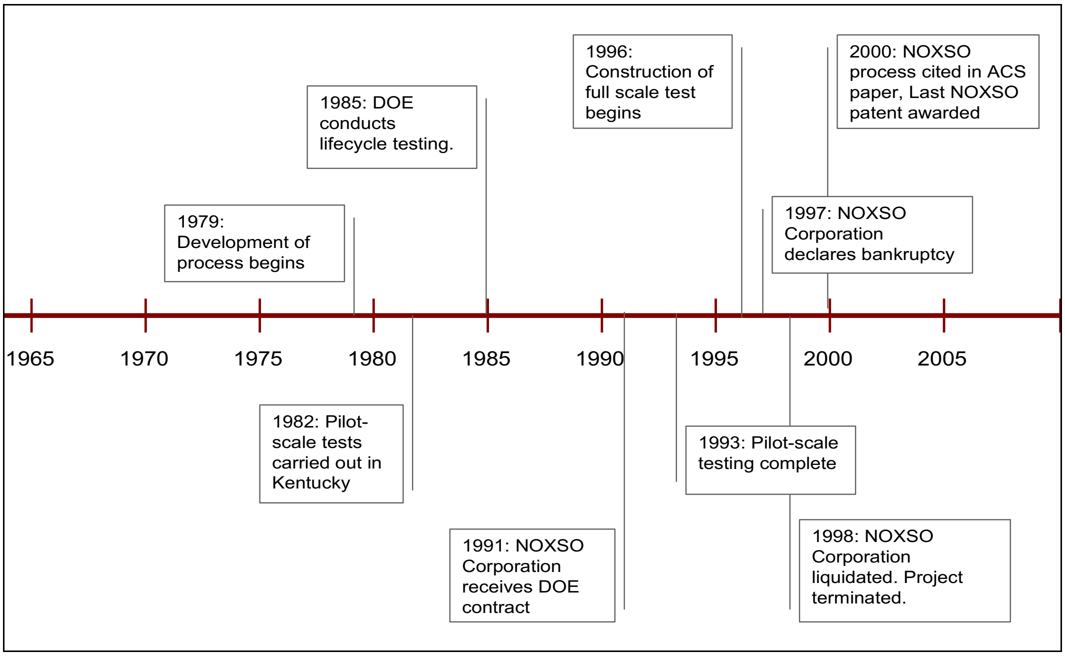

Fig. 38. Development history of the NOXSO process for post-combustion SO_2 and NO_x capture.

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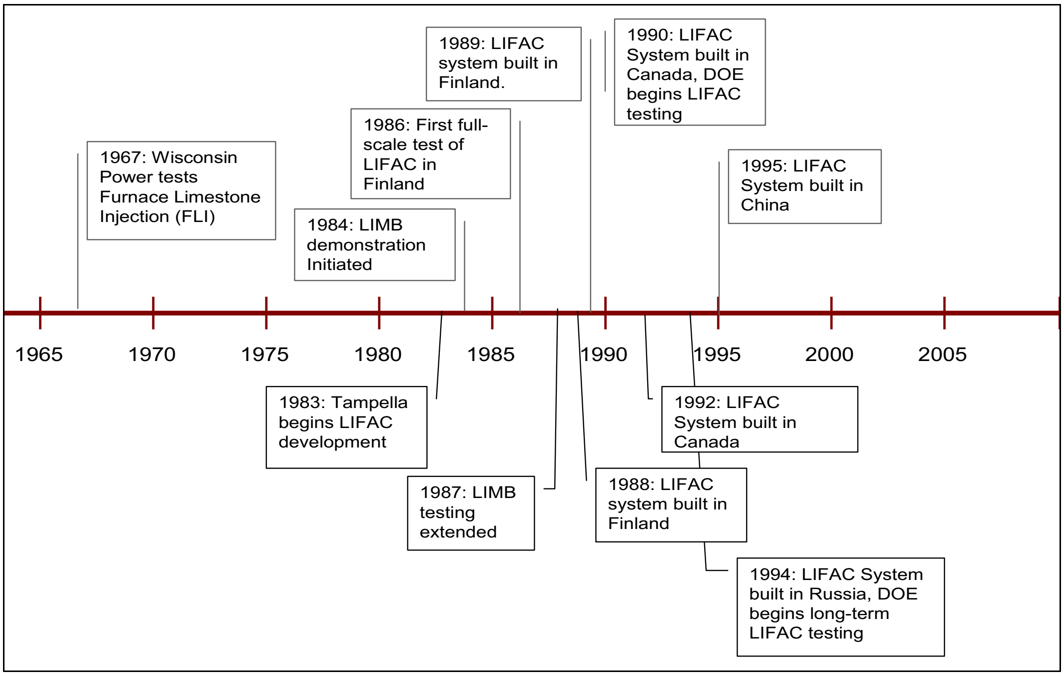

Fig. 39. Development history of the furnace limestone injection process for SO_2 capture.

capture systems today, at the time they were being developed there were no requirements for (hence, no significant markets for) high-efficiency combined SO_2-NO_x capture systems, or moderately efficient SO_2 removal systems. This factor alone posed high risks for their commercial success. While this was consistent with the DOE mission of pursuing high risk, high payoff technologies, the high payoffs that were projected never materialized—in large part because the markets for these technologies failed to develop as expected. Similar risks face advanced carbon capture technologies today.

Figs. 37–40 also show that the time required to develop a novel capture process from concept to large-scale demonstration was typically two to three decades. During this period the projected economic benefits of the advanced technologies tended to shrink. Not only did their cost tend to rise during the development process (as suggested earlier in Fig. 27), but the cost of competing options also fell. Thus, the continual deployment and improvement of commercial FGD systems (mainly in the U.S.) and SCR systems (in Japan and Germany) during the 1980s made it increasingly difficult for combined SO_2-NO_x capture technologies to gain a foothold and

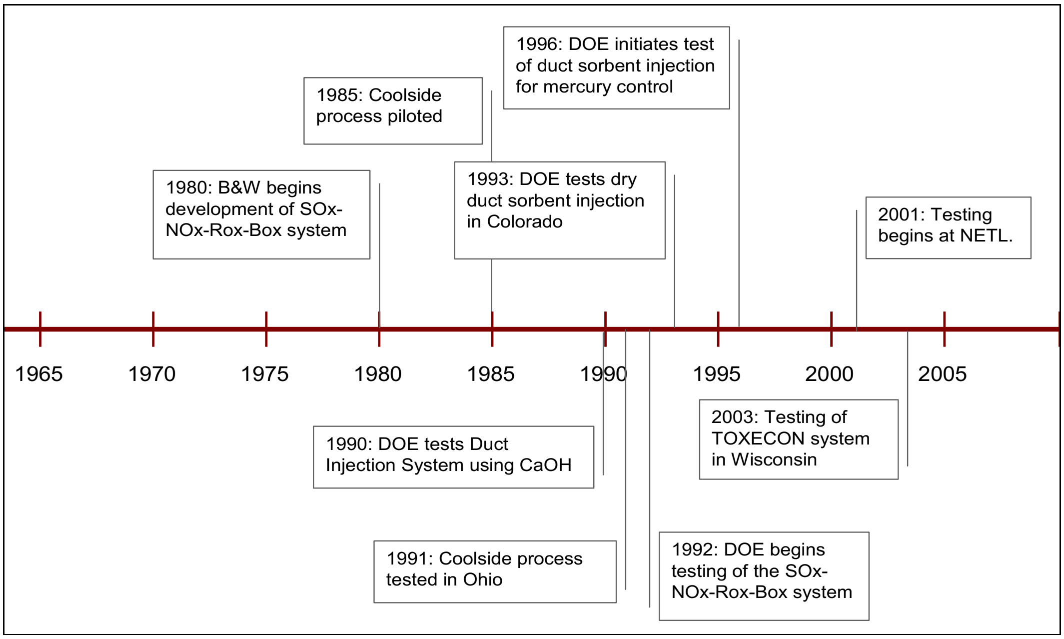

Fig. 40. Development history of the duct sorbent injection process for SO_2 capture.

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compete in the marketplace. Indeed, in the U.S., there was no market for post-combustion NO_x capture at coal-burning plants until the mid-1990s [106]. In the case of furnace and duct sorbent injection processes for moderate levels of SO_2 capture, the anticipated market for such an option did materialize. In the U.S., the acid rain provisions of the 1990 Clean Air Act Amendments introduced a cap-and-trade program for SO_2 that provided power plant operators with greater flexibility in meeting emission reduction requirements. Switching to low-sulfur coal proved to be an easier and more economical choice than sorbent injection, especially as low-sulfur western coals entered the marketplace.

In terms of lessons learned, the above discussion suggests that the lengthy time historically required to develop advanced environmental technologies tends to diminish the probability of commercial success, as more mature technologies gain initial market share (assuming the existence of a market). Thus, any efforts that can accelerate the development and commercialization of new technology can help reduce the commercial risks. Apropos of that, another lesson drawn from this experience is that current commercial technologies do not “stand still”—as is often assumed by proponents and developers of new technologies. Improvements and cost reductions in current systems also must be anticipated to realistically assess the prospects and potential payoffs of an advanced technology that is still under development.

7.3. *The pace of capture technology development*

Historical deployment rates for power plant environmental technologies also can serve as a useful guide for realistically assessing current projections for CO_2 capture technology. Fig. 41 shows the trends in deployment of post-combustion capture systems for SO_2 and NO_x in different countries from 1970 to 2000. For FGD systems, the maximum rate of deployment in response to new environmental policy requirements over this period was approximately 15 GW per year (in Germany), with an average rate of about 8 GW per year worldwide. For SCR systems, the maximum rate was about 10 GW per year (again in Germany), with an average global deployment rate of about 5 GW per year. These results suggests that deployment scenarios for CO_2 capture systems that significantly exceed these rate may be unrealistic or will require aggressive new efforts and measures to achieve.

7.4. *Rates of performance and cost improvements*

Studies also have documented the historical rates of improvement in the performance (capture efficiency) of power plant emission control systems and their rates of cost reduction following commercialization [82,128]. For example, Fig. 42 shows the trend in average SO_2 capture efficiency for power plant FGD systems coming online in the U.S. from 1969 to 1995. Capture efficiencies increased from about 70 percent to 95 percent over that period due to the combined effects of technology improvements and regulatory requirements. Since that time the performance of wet FGD systems has continued to improve, with new systems today capturing 98 to 99 percent or more of the SO_2. These deep levels of sulfur removal capability can now facilitate post-combustion CO_2 capture since very low inlet SO_2 concentrations are required for current amine-based systems [129] as well as for most of the advanced processes under

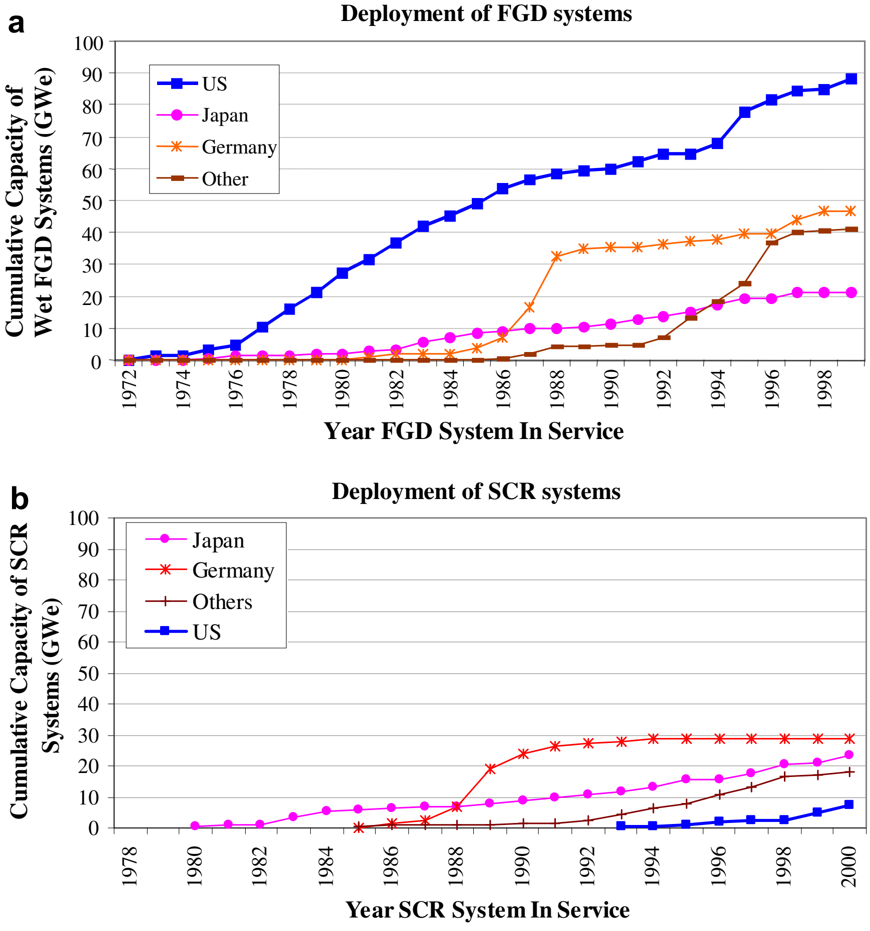

Fig. 41. Historical deployment for post-combustion SO_2 and NO_x capture systems (FGD and SCR technologies) [82].

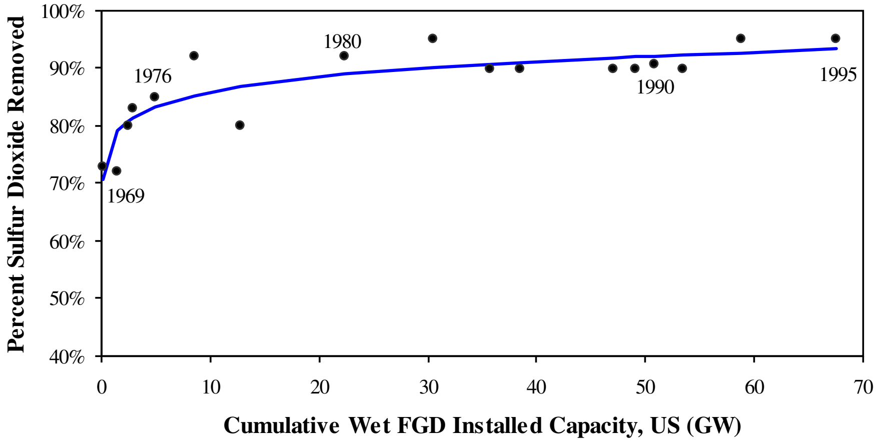

Fig. 42. Improvements in SO_2 removal efficiency of commercial lime and limestone FGD systems coming online in a given year, as a function of cumulative installed FGD capacity in the U.S [98].

development. Historical trends of increasing efficiency of post-combustion capture systems for power plant particulate and NO_x emissions [128] offer further indications that CO_2 capture systems also have the potential for future performance improvements.

Regarding costs, Fig. 43 shows the historical trends in capital costs for FGD and SCR systems for a standardized coal-fired power plant design. In both cases, the actual or estimated capital cost (as well as O&M costs) increased during the early commercialization of these technologies in order to achieve the levels of availability and performance required for utility operations. Subsequently, costs declined considerably with increasing deployment. On average, the capital cost of these technologies fell by about 13 percent for each doubling of total installed capacity [17]. This historical "learning rate" was assumed for future CO_2 capture systems in the plant-level cost projections shown earlier in Fig. 30, after allowing for a period in which costs might rise during early commercialization (as suggested in Fig. 43).

7.5. The critical role of government actions

In the U.S. economy, the existence of a market (demand) for a product is critical to its adoption and widespread use. This is true as well for CO_2 capture technologies. The adoption and diffusion of a technology also are key elements of the innovation process that improves the performance of a product and reduces its cost over time, as depicted earlier in Fig. 27. R&D plays a critical role in this process. But R&D alone is not sufficient without a market for the technology.

For environmental technologies such as CO_2 capture and storage systems, few if any markets exist in the absence of government actions and policies. What electric utility company, for example, would want to spend a large sum of money to install CCS—even with an improved lower-cost capture process—if there is no requirement or incentive to reduce CO_2 emissions? A costly action such as this provides little or no economic value to the company—indeed, the added cost and energy penalty of CO_2 capture increase the cost of operation. Only if a government action either requires CO_2 capture and storage, or makes it financially attractive, would a sizeable market for capture technologies develop. Thus, as with other environmental pollutants that affect the public welfare, government actions are needed to create or enhance markets for CO_2 emission-reducing technologies.

Different policy measures influence markets in different ways. Measures such as government loan guarantees, tax credits, direct financial subsides and R&D funding can help create markets by providing incentives for technology development, deployment and diffusion. Voluntary incentives such as these are commonly referred to as "technology policy" measures [130]. In contrast, regulatory policies such as an emissions cap, emissions tax, or performance standards that limit emissions to specified levels are

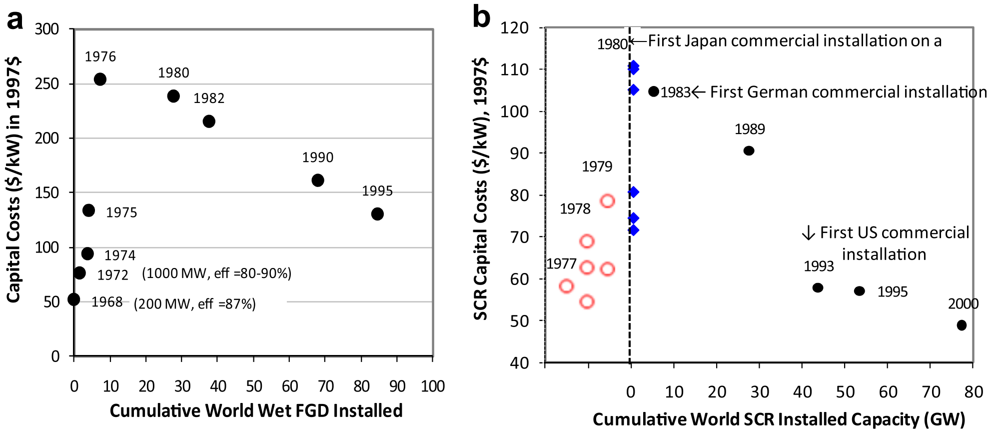

Fig. 43. (a) Capital cost trends for a wet limestone FGD system at a standardized new power plant (500 MW, 3.5% sulfur coal, 90% SO_2 removal, except where noted); and, (b) capital cost trends for an SCR system at a new plant (500 MW, medium sulfur coal, 80% NO_x removal). Solid diamond symbols are studies based on low-sulfur coal plants. Open circles are studies prior to SCR use on coal-fired power plants [82].

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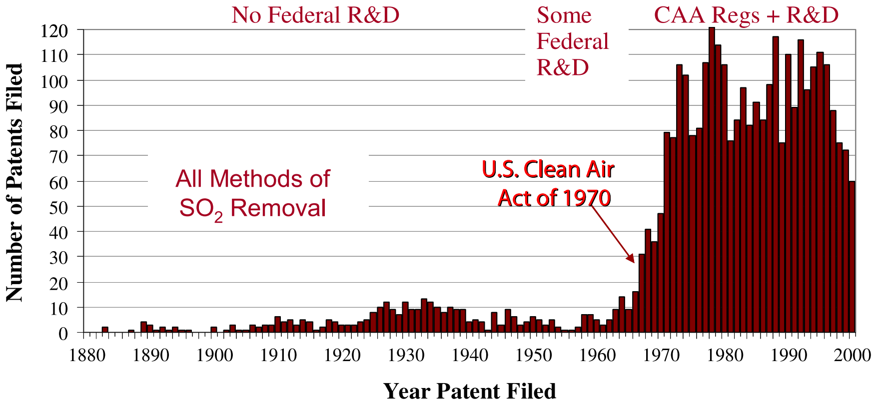

Fig. 44. Trend in U.S. patenting activity for SO_2 removal technologies [131].

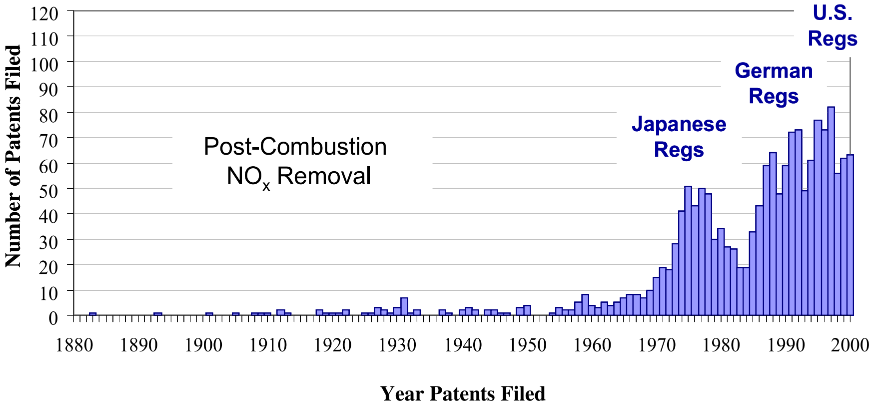

Fig. 45. Trend in U.S. patenting activity for post-combustion NO_x removal technologies [111].

mandatory, not voluntary. These policies create or expand markets for lower-emission technologies by imposing requirements that can be met only, or most economically, with a low emission technology.

Through its influence on markets for environmental technologies like CO_2 capture and storage systems, government actions are thus a critical element of the technological innovation process. Studies of past measures to reduce sulfur dioxide and nitrogen oxide emissions from U.S. power plants have documented the ability of regulatory policies to influence both the magnitude and direction of R&D efforts to develop new or improved capture technologies [111,131]. Figs. 44 and 45, for example, show the century-scale trends in U.S. patenting activity for SO_2 reduction technologies and post-combustion NO_x removal systems, respectively. In both cases, the number of new patents filed—a measure of “inventive activity”—increased dramatically when new environmental regulations that required or incentivized the use of these technologies was anticipated and came into force. In the case of post-combustion NO_x controls, such regulations for coal plants materialized first in Japan and then in Germany; similar regulations in the U.S. lagged by more than a decade. The subsequent reduction in cost that accompanied the increased deployment of these technologies (Fig. 45) is evidence of the influence of government policies on technology innovation in this domain.

7.6. Conclusions from past experience

The analysis of recent historical experience with the U.S. R&D program to develop advanced lower-cost technologies for post-combustion capture of sulfur dioxide and nitrogen oxides at coal-fired power plants revealed that several decades was commonly required to bring a new process concept to commercial scale demonstration. The lack of commercial success in most of these ventures also illustrated the risks inherent in developing new environmental technologies, especially when there is not yet a significant market demand for them. Benchmark rates of technology deployment and long-term cost reductions for commercially successful capture technologies also were derived from U.S. and global experience with FGD systems (for SO_2 capture) and SCR systems (for NO_x capture). These historical data underscore the ambitious and challenging nature of current roadmaps and projections for the commercialization and deployments of advanced CO_2 capture processes in the absence of a strong policy driver for carbon emission reductions.

8. Summary and conclusions

This paper has sought to provide a realistic assessment of prospects for improved, lower-cost CO_2 capture systems for use at power plants and other industrial facilities to mitigate emissions of greenhouse gases linked to global climate change. Toward that end, the paper first described each of the three current approaches to CO_2 capture, namely, post-combustion capture from power plant flue gases using solvents such as MEA or other amines; pre-combustion capture, also via chemical solvents, from the synthesis gas of an integrated coal gasification combined cycle power plant; and oxy-combustion capture, in which high-purity oxygen is used for combustion to produce a flue gas with high CO_2 concentrations amenable to capture without a chemical process.

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Currently, post-combustion and pre-combustion capture technologies are commercial and widely used for gas stream purification in a variety of industrial processes. CO_2 is also captured from the flue gases of several small-scale power plant installations (to produce commodity CO_2 for sale), but has not yet been demonstrated at full-scale for coal or gas-powered power plants. Oxy-combustion capture is still under development and is not yet commercial, but is advancing rapidly as an option for power plant CO_2 capture. The advantages and limitations of each of these three methods were discussed, along with plans for their development and demonstration in large-scale power plant applications.

While all three approaches are capable of high CO_2 capture efficiencies (typically about 90 percent), major drawbacks of current capture processes are their high cost per unit of power produced. The large energy requirements for CO_2 capture (roughly 15%–30% more energy needed per net kWh for new fossil fuel plants) contribute significantly to this high cost since larger plants must be built to supply a given net power demand. This is especially true for combustion-based power plants, where CO_2 capture incurs higher energy penalties and higher additional costs than at gasification-based plants. Retrofits of CO_2 capture systems to existing power plants also tend to be more costly on a per kilowatt-hour basis than capture at new plants, in large part because the low plant efficiency typical of existing units results in a large reduction in net power output.

Substantial R&D activities are underway worldwide to develop improved solvents for post-combustion capture, as well as potential "breakthrough technologies" such as novel solvents, sorbents, membranes and oxyfuel systems that hold promise for lower-cost capture technology. Most of these processes are still in the early stages of research and development, so that credible estimates of their performance and (especially) cost are lacking at this time. Even with an aggressive development schedule, the commercial availability of these advanced technologies is at least a decade away, and likely longer based on past experience with other technology development programs.

Capture processes that are undergoing testing and evaluation at the large pilot plant scale are, for the most part, new or improved solvent formulations such as ammonia and advanced amines. These new solvents could be available for commercial use in post-combustion systems within several years if subsequent full-scale testing confirms their overall benefit. Pilot-scale oxy-combustion tests also are now underway at several international locations, while two European IGCC power plants recently installed slip stream pilot plants to evaluate new pre-combustion capture options.

As of the writing there are still no full-scale applications of CO_2 capture at a coal-based or gas-fired power plant. However, a number of large-scale demonstration projects for post-, pre- and oxy-combustion systems are planned or underway in several countries. Large-scale capture projects also are planned at several types of industrial facilities.

In general, the focus of most current R&D is on cost reduction as opposed to gains in the efficiency of CO_2 capture. While many R&D programs emphasize the need for lower-cost retrofit technologies suitable for existing power plants, as a practical matter most advanced technologies are benchmarked on (and best suited for) new plant or repowering applications.

Whether for new power plants or existing units, the key questions are: when will advanced CO_2 capture systems be available for commercial roll-out? And how much cheaper will they be compared to current technology? The technology roadmaps reviewed in this paper anticipate that CO_2 capture will be available for commercial deployment at power plants by 2020. For current commercial technologies like post-combustion amine systems, this is a conservative estimate since the key requirement is for scale-up and demonstration at a full-size power plant—achievable well before 2020. A number of roadmaps project that novel technology like solid sorbent systems for post-combustion capture also will be commercial in the 2020 time frame. Such projections acknowledge, however, that this will require aggressive and sustained efforts to advance promising concepts to commercial reality.

That caveat is supported by our review of past experience from R&D programs to develop lower-cost technologies for post-combustion SO_2 and NO_x capture at coal-fired power plants. Those efforts typically took two to three decades to bring new concepts like a combined SO_2 and NO_x capture system to commercial availability. In most cases, however, the cost advantage initially foreseen largely evaporated over this period: the advanced technology tended to get more expensive as the development process progressed (consistent with "textbook" descriptions of the innovation process), while the cost of existing commercial options gradually declined over time. The absence of a market for these advanced technologies during their development stage put them at a further disadvantage since there was little incentive for their commercialization and large-scale deployment.

The good news based on past experience is that the cost of environmental technologies that succeed in the marketplace tends to fall over time. For example, after an initial rise during the early commercialization period, the cost of post-combustion SO_2 and NO_x capture systems declined by 50 percent or more after about two decades of deployment at coal-fired power plants. This trend is consistent with the "learning curve" behavior seen for many other classes of technology. It appears reasonable to expect a similar trend for future CO_2 capture costs once a market is established that encourages capture technologies to become widely deployed.

This paper also showed that the cost of CO_2 capture depends strongly on other aspects of power plant design, financing and operation—not solely on the cost of the CO_2 capture unit. Future improvements in power plant efficiency, for example, also will tend to lower the unit cost of CO_2 capture and must therefore also be a focus of long-term R&D.

This review also noted that published estimates of future electricity costs for advanced power plants and CO_2 capture systems offer optimistic projections of cost reductions relative to current systems. In general, the further away a technology is from commercial reality, the better it tends to look. Historically, however, estimates of technology cost tend to rise along the path to commercialization. Thus, there is considerably uncertainty in the projected cost of technologies that are not yet commercial, especially those that exist only as conceptual designs.

More reliable estimates of future technology costs typically are linked to projections of their expected level of commercial deployment in a given time frame—i.e., a measure of their market size. For power plant technologies this is commonly expressed as total installed capacity. For technologies like CO_2 capture systems, whose sole purpose is to control environmental emissions, there is no significant market in the absence of government actions or policies that effectively create such markets—either through regulations that limit CO_2 emissions or through voluntary incentives for its use. The historical evidence and technical literature examined in this paper strongly link future cost reductions to the level of commercial deployment of a technology. In empirical "experience curve" models, the plant capacity in service serves as a surrogate for the many factors that influence its future cost, including expenditures for R&D and the knowledge gained through learning-by-doing (related to manufacturing) and learning-by-using (related to technology use).

Based on such models, published estimates project the future cost of electricity from power plants with CO_2 capture to fall by up to 30 percent below current levels after roughly 100,000 MW of capture plant capacity has been installed and operated worldwide. That would represent a significant decrease from current costs—one that would bring the cost and efficiency of future power plants with CO_2 capture close to that of current plants without capture. (For reference, it took approximately twenty years to deploy 100,000 MW of SO_2 capture systems worldwide, following passage of the U.S. 1970 Clean Air Act Amendments.) Uncertainty estimates, however, indicate that future CO_2 capture cost reductions could be smaller or larger than indicated above. Thus, whether future cost reductions will meet, exceed or fall short of current estimates will only be known with hindsight. Similarly, only hindsight will tell the extent to which future lower-cost capture technologies will include systems currently in the R&D pipeline.

In the context of this paper, the key insight governing realistic prospects for improved carbon capture technology is that achieving significant cost reductions will require not only a vigorous and sustained level of R&D, as is currently underway, but also a substantial level of commercial deployment. That, in turn, will require a significant market for CO_2 capture technologies that can only be established by government actions. At present such a market does not exist. While various types of incentive programs can accelerate the development and deployment of CO_2 capture technology, actions that significantly limit emissions of CO_2 to the atmosphere ultimately are needed to realize substantial and sustained reductions in the future cost of CO_2 capture.

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