An air–liquid contactor for large-scale capture of CO_2 from air

By Geoffrey Holmes[1] and David W. Keith[1,2,*]

[1]*Carbon Engineering Ltd, EEEL467, 2500 University Drive NW, Calgary, Alberta, Canada T2N 1N4*
[2]*School of Engineering and Applied Sciences, and Kennedy School of Government, Harvard University, 29 Oxford Street, Cambridge, MA 02138, USA*

We present a conceptually simple method for optimizing the design of a gas–liquid contactor for capture of carbon dioxide from ambient air, or 'air capture'. We apply the method to a slab geometry contactor that uses components, design and fabrication methods derived from cooling towers. We use mass transfer data appropriate for capture using a strong NaOH solution, combined with engineering and cost data derived from engineering studies performed by Carbon Engineering Ltd, and find that the total costs for air contacting alone—no regeneration—can be of the order of $60 per tonne CO_2. We analyse the reasons why our cost estimate diverges from that of other recent reports and conclude that the divergence arises from fundamental design choices rather than from differences in costing methodology. Finally, we review the technology risks and conclude that they can be readily addressed by prototype testing.

Keywords: air capture; carbon dioxide removal; cost optimization

1. Introduction

Air capture (AC) is a technology for capturing carbon dioxide from ambient air in a closed-loop industrial process. AC is one of an emerging set of technologies for carbon dioxide removal, which includes geological storage of biotic carbon and the acceleration of geochemical weathering [1]. Capture of CO_2 from air has been commercialized as a pre-treatment before cryogenic air separation. It is possible to design industrial-scale AC systems using current commercial processes in a problem-specific design, but no commercial-scale AC system has yet been realized. Arguments about the wisdom of investing in the development of AC technology turn on estimates of its cost, and of the potential for cost reductions, both of which are hotly disputed. A recent report by the American Physical Society (APS) concludes that AC would cost roughly $600–800 per tonne CO_2 using current technologies and suggests that there is little prospect for substantial

near-term improvement in these costs [2]. A recent analysis by House *et al.* [3] concluded that the cost would be about \$1000 per tonne CO_2. Our engineering analysis suggests that overall cost will be much lower. Here, our focus is limited to the design and costing of a gas–liquid contactor for AC. While we do not analyse an end-to-end AC system, our results are relevant to the full system analysis because contactor costs account for over half of the APS cost estimate, and we demonstrate here that contactor cost can be roughly a factor of four lower, using an alternative design with the same costing methodology.

In this study, we develop a simplified method for optimizing the design of a liquid–gas contactor for capturing CO_2 from air in strong hydroxide solutions, and provide cost and performance data sufficient to enable first-order evaluation of contactor cost. We show that costs for contacting alone can be below \$60 per tonne CO_2. A complete AC system requires both a contactor and a system for regenerating the absorbing solution. We do not provide any new analysis of the regeneration cycle here. Finally, this study examines the differences between our estimate and that provided by the APS. We find that the fourfold discrepancy in cost estimates for the contactor arise not from differences in costing methodology, but from the APS's choice of a design, which, while low-risk, is ill-suited to the task.

If it can be developed commercially, AC will occupy a technological niche quite distinct from traditional point-source carbon capture and storage (CCS). AC will cost more than CCS when both are operated under the same economic conditions, though it may, in some circumstances, compete with post-combustion capture at power plants because it enables one to exploit the global heterogeneity in costs of energy, labour and CO_2 disposal. For example, an AC facility operating on low-cost 'stranded' natural gas that is able to provide CO_2 for enhanced oil recovery at a location without other CO_2 sources might be competitive with post-combustion capture in high-cost locations such as Canadian oil sands operations.

The central advantages of AC, should it be realized at cost-effective industrial scale, are as follows. (i) It allows one to apply industrial economies of scale to small and mobile emission sources—about 60 per cent of global carbon emissions—which enables a partial decoupling of carbon capture from the energy infrastructure. (ii) It provides a route to managing carbon emissions in the transportation sector—where carbon mitigation costs are generally higher than in the electric sector—by enabling the generation of carbon-neutral hydrocarbons [4]. We note that, without AC, prospects for deep reductions in emissions from transportation are more limited than elsewhere in the economy. Electric vehicles show great promise but their ability to make deep reductions across the entire transportation sector is uncertain, as is the potential for large-scale hydrogen and environmentally compatible biofuels. (iii) Finally, while humanity has many options for reducing emissions, none but carbon removal and solar geoengineering enable significant reduction in the overall climate risk, because risks grow with the accumulating stock of atmospheric CO_2, which cannot be reduced without industrial-scale carbon-negative technologies such as AC. Elimination of emissions stops the increase in the climate risk but cannot meaningfully reduce it on a time scale shorter than a few centuries. Together, these advantages may outweigh the intrinsic difficulty of capturing carbon from the air and mean that AC may be important even if it is not competitive with CCS at large point sources.
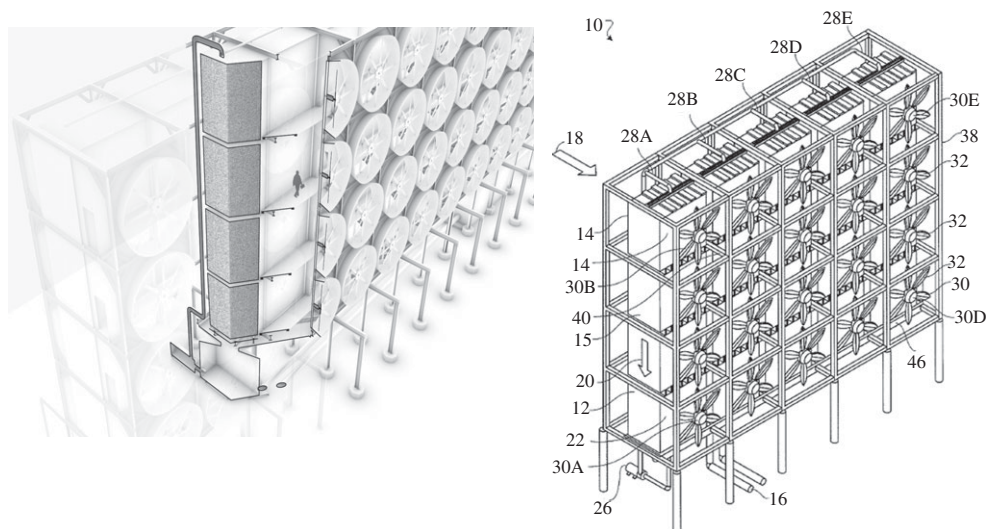
Figure 1. Carbon Engineering slab air-contactor design.

It is important to note that most of the work reported here has been performed within Carbon Engineering Ltd (CE), an independent company (based in Calgary, Canada) that is engineering industrial-scale AC technology. The design data presented here are based on the performance of commercially available structured packing and on previously published estimates of the mass transfer of CO_2 into strong hydroxide solutions. New data from an independent engineering study of our contactor design are presented here, along with CE's laboratory data, which confirm these performance estimates when commercial packings are used with NaOH and KOH solutions in laboratory-scale tests.

2. Liquid-based packed slab air-contactor

Air contacting for direct AC has commonalities with existing gas separation technologies, and also with large-scale cooling towers. Like an industrial cooling unit, AC requires ingestion of very large quantities of ambient air, and management of ingested debris and of droplets, 'drift', that might be released to the environment. Our contactor engineering has focused on large forced-draught units with cross-flow slab geometry, where *cross-flow* means that the air flow is orthogonal to the gravity-driven downward flow of the liquid and *slab geometry* means that the overall contactor is thin along the air flow axis compared with its height and length. A representation of CE's slab contactor design is shown in figure 1.

Here, we present a simplified cost-optimization model that describes the engineering constraints and cost trade-offs at the heart of our contactor design. We provide a rationale for the fundamental design choices mentioned earlier. The cost model is presented in §2*a* and the justification for the most important cost and performance parameters is given in §2*b–e*. The results of the cost model are described in §2*f*. We close with a discussion of technology risks and uncertainties in §2*g*.

As mentioned in the introductory section, the exact composition of the absorbing fluid is a design choice that is determined by co-optimization of the contactor and regeneration system. The design presented here assumes that absorber fluid is an aqueous solution that absorbs CO_2 from ambient air with flux across the surface of the liquid film of order $1 \, \text{mg m}^{-2} \, \text{s}^{-1}$ (see §2e for rationale), and that, under typical operating conditions, each kilogram of solution absorbs about $20 \, \text{g}$ of CO_2 before it is returned for regeneration. These properties are typical of a 1–2 M NaOH solution.

Under these conditions, the theoretical liquid-to-gas volumetric flow ratio required in the contactor is about $1 : 50\,000$. This ratio is much smaller—less liquid per unit gas—than is typically seen in liquid–gas contactors used in chemical engineering applications. This gives us the flexibility to operate the packing in a lower liquid flow regime than is common in chemical scrubbers and in cooling towers. The advantage of low liquid flow rates is that liquid pumping can be reduced to a minor factor driving system configuration and cost; while the disadvantage is that low flows increase the difficulty of keeping the packing evenly wetted. We maintain wetting at low average flow rates by varying the flow, alternating short-period high flow rates that wet the packing and remove dust and debris with low flow rates that replenish the wetted surface. The manipulation of flow rates to maintain wetting in these configurations, described in §2d, is an important part of the CE contactor design.

Economies of scale in the regeneration system drive us to systems that capture CO_2 at a rate of the order of 0.1–1 megaton per year (Mt yr^{-1}). We do not present the overall system scale trade-off here; rather, we will simply assume that the contactor must capture $1 \, \text{Mt yr}^{-1}$ and discuss the design of such a system. At 400 ppm, air is 0.06 per cent CO_2 by weight; so a contactor with a $1 \, \text{Mt yr}^{-1}$ capture rate must process at least $46\,000 \, \text{m}^3 \, \text{s}^{-1}$ of air ($1300 \, \text{km}^3 \, \text{yr}^{-1}$). The choice of air velocity is one of the many trade-offs between capital and operating costs. High air velocities increase energy consumption, which is a primary contributor to operating costs, but they also minimize the required contactor inlet area, and hence reduce the contribution of capital cost to each unit of CO_2 captured. The results of the cost optimization described below show that optimal air velocities are in the range 1–3 m s^{-1}.

Note that while AC literature has often mentioned the use of ambient winds, our engineering has shown that the capital expenditure of equipment contributes a large enough fraction of the overall capture cost that forced-draught air flow is essential to avoid 'down-time' when winds are too low and to allow continuous operation of the whole process.

Given an air velocity of $1.5 \, \text{m s}^{-1}$, the required inlet area is $38\,000 \, \text{m}^2$ to capture $1 \, \text{Mt yr}^{-1}$, assuming a CO_2 capture fraction of 0.75 (representative of the optimal capture fractions, as we will show below). Perhaps the most conservative design would be to use multiple packed towers. The advantage of this system is that the use of packed towers for gas–liquid exchange is a standard component of the worldwide chemical industry. This system has no technical risk because such packed towers were widely used in commercial systems that removed CO_2 from air with KOH solutions as a pre-treatment for cryogenic air separation. Packed towers have several serious disadvantages as a basis for a large-scale AC contactor. The largest packed towers have inlet areas of about $100 \, \text{m}^2$; so hundreds would be required to capture $1 \, \text{Mt yr}^{-1}$. An array of these towers lacks a mechanism to

avoid recycling of the low-CO_2 outlet air from one unit into the inlet of another, and yet such recycling must be minimized in a cost-effective system. Moreover, the multiple tower geometry is far more expensive because of the cost versus scale relationships and the complexity of the required piping connections.

The alternative is to adapt technology used in large-scale cooling towers and waste treatment facilities, which are designed to efficiently bring very large quantities of ambient air into contact with fluids. As described in §2*b*, our slab geometry design is based closely on the design experience in these industries. There are technical risks associated with adapting cooling tower technology to the chemical process used in AC; we address these risks and the steps we are taking to minimize them in §2*g*.

The engineering study described in §2*b* arrived at an optimized air-contactor design that is roughly 20 m tall, 8 m deep and 200 m long. In CE's full-scale facility design, roughly 10 contacting units would be dispersed around a central regeneration, compression and processing facility, to cumulatively capture 1 Mt yr^{-1}.

(*a*) Air-contactor cost optimization model

The simplified cost model assumes that the contactor's capital cost can be divided into two independent components, the shell and the packing. The *packing* cost, C_P, is proportional to the packing volume and it includes the installed cost of the packing itself along with the specialized supports, fluid distributors and drift eliminators. The *shell* includes the structure, fans, fluid pumps and large-scale fluid piping (but not the distributors themselves, which are included in C_P). We assume that the *shell* cost, C_A, is proportional to the frontal swept area of the packing. As we describe in §2*b*, these costs are full owners' cost, including site acquisition, all construction and engineering costs as well as interest during construction.

The core simplification of this cost model is that, for a slab geometry contactor, the *shell* cost is roughly independent of the depth of the packing. In part, this is because maximum wind loads play a larger role in determining slab structural design than does the weight of the packing. For example, the cost of a contactor shell with a 20 × 200 m frontal area does not change significantly as the thickness of the packing is varied from 3 to 15 m.

As discussed in §2*e*, the CO_2 mass flux into the liquid is always proportional to the concentration of CO_2 in the overlying air so that it can be expressed as a liquid-phase mass transfer coefficient K_L (with units of velocity) times the CO_2 mass density in the air [5].

The complete model equations are shown in table 1, with variables defined in table 2. Here, we provide a few notes of explanation and interpretation.

The CO_2 captured per unit inlet area of the slab contactor is expressed as a mass flux (equation (2.1)). The airborne CO_2 concentration decays exponentially through the packing depth with a length scale of $V/(\varepsilon \, SSA \, K_L)$, which arises from the ratio of the gas velocity to the CO_2 removal flux, which is the mass transfer coefficient times the surface area per unit volume. The packing efficiency, ε, accounts for imperfect wetting of the packing area. Note that equation (2.1) is a simple and robust approximation; in reality, hydroxide concentration and hence uptake rate vary within the packing. We have constructed a full two-dimensional

Table 1. Cost optimization model equations.

equation	name	formula	units
(2.1)	CO_2 capture flux	$F = f_{op}\rho_{CO_2} V(1 - e^{-\varepsilon\,\text{SSA}\,D\,K_L/V})$	$\text{kg m}^{-2}\text{yr}^{-1}$
(2.2)	pressure drop in packing[a]	$\Delta P = D7.4\,V^{2.14}$	Pa
(2.3)	energy for fans	$E = f_{op}\Delta PV/\eta_{fan}$	$\text{J m}^{-2}\,\text{yr}^{-1}$
(2.4)	capital cost	$C_{capital} = C_A + C_{pack}D$	$\$\,\text{m}^{-2}$
(2.5)	operating cost	$C_{operating} = EC_{elec} + \text{M\&O}\,C_{capital}$	$\$\,\text{m}^{-2}\,\text{yr}^{-1}$
(2.6)	total cost minimization	$\min_{V,D} C_{CO_2} = (C_{operating} + \text{CCF}\,C_{capital})/F$	$\$$ per tonne CO_2

[a]Vendor-specified pressure drop for Brentwood Industries XF12560 packing, simplified to low liquid flow scenario.

Table 2. Cost optimization variables, values and units.

variable	units	value	notes and typical values
η_{fan}	%	56	fan efficiency (§2c)
C_{elec}	$\$\,\text{J}^{-1}$	2.2×10^{-8}	cost of electricity ($\$80$ per MW h)
C_A	$\$\,\text{m}^{-2}$	3700	capital cost per frontal area (§2b)
C_{pack}	$\$\,\text{m}^{-3}$	250	packing and fluid distributor cost (§2d)
f_{op}	s per year	2.7×10^7	assumed annual operation fraction (85%)
CCF	% per year	15	capital charge factor
M\&O	% per year	5	maintenance and operation
ε	%	80	packing efficiency (§2d)
ρ_{CO_2}	kg m^{-3}	7.3×10^{-4}	mass density of CO_2 in air at 400 ppmv
SSA	$\text{m}^2\,\text{m}^{-3}$	210	specific packing area (§2d)
K_L	m s^{-1}	1.5×10^{-3}	mass transfer coefficient (§2e)
V	m s^{-1}	1–2	air velocity (optimal values computed in §2f)
D	m	5–15	packing depth (optimal values computed in §2f)

model as an Excel module in Aspen Plus that accounts for gas flow and depletion of the hydroxide in the liquid in contact with intermittent fluid flow, and we have tested the model in various experimental geometries.

The energy cost of operating the contactor is simply determined from the pressure drop (equation (2.2)) and the pressure–volume work term (equation (2.3)).

To simplify life-cycle analysis, all of our full system designs supply all electricity needs with on-site generation that is thermally integrated with the regeneration cycle. Almost all CO_2 from the process is captured as part of the regeneration cycle. In this simplified contactor-only model, we simply assume a cost of electricity of $\$80$ per MW h. In the full design, the two systems must, of course, be optimized jointly.

We adopt 15 per cent as an overall annual capital charge factor, which includes interest, depreciation and taxes. The annual cost of operating personnel and regular maintenance is estimated annually as 5 per cent of the system capital cost.

Table 3. CE 1 Mt yr^{-1} contactor array, structural cost analysis breakdown.

item	labour[a] (%)	materials (%)	percentage of cost	capital cost (C$ million)
fans	3.8	14.8	18.7	26.3
packing	1.4	17.4	18.9	26.6
pumps	2.6	1.0	3.5	4.9
structure	27.7	8.5	36.2	50.9
admin/contingency			22.7	31.9
total	35.5	41.8	100	140.6

[a]Labour costs include all project management costs as these costs were allocated in proportion to labour costs.

Finally, the loss of water through evaporation results in the need for a make-up water stream priced at an average [6] of $0.5 per tonne H_2O, and thus adds of the order of $5 to the cost of every tonne of captured CO_2. For simplicity in the rest of our analysis, we ignore this cost.

(b) Contactor capital cost

Our estimate of the cost per unit contactor inlet area, C_A, derives from a design study performed by CE and a sub-contractor, on a purpose-built air-capture contactor, and from cost estimates for industrial cooling tower hardware.

CE contracted the engineering, procurement and construction firm SolTech Projects Inc. (Calgary, Canada) in 2009 to develop an engineering design based upon CE's slab air-contactor concept. In this initial study, CE specified packing performance, including weight, liquid flow rate requirements and pressure drop, and SolTech was responsible for design and costing of only the shell around the packing. SolTech performed a full structural engineering analysis and simulated 100-year event wind and snow loading scenarios and examined overall cost sensitivity to the height, width and length of the contactor modules. CE's current 20×200 m module size is a result of this structural cost optimization.

SolTech's 'bottom-up' cost analysis and engineering included vendor quotes for site preparations, foundation, structural materials and construction, field assembly, air and liquid handling systems, as well as standard project management and contingency. In addition, SolTech performed an independent 'top-down' factor and scaling analysis of system cost to assess the accuracy of the cost analysis and to account for items not included in this first design study.

The full, installed cost of a single 20×200 m (note that these are packing dimensions, external size is larger) CE slab air-contactor module was C$15 million (Canadian dollars). A top-level cost breakdown is presented in table 3.

Note that cost estimation was based on 2008–2009, Alberta-specific prices and labour rates, which were among the highest in the world due to rapid growth in the Alberta petroleum industry. These Alberta costs are typically 30–50% higher that US Gulf Coast costs. Note that worldwide construction cost indices peaked in 2008. For example, the IHS-CERA 'Downstream' capital cost index increased 85 per cent from 2003 to its 2008 peak, from which it has now declined about 20 per cent.

Table 4. Structural cost comparisons—cooling towers, CE slab contactor, traditional packed tower.

	modular counter-flow cooling unit	large cross-flow cooling tower	CE slab design (single module)	packed tower
inlet area (m^2)	30	1750	4000	113
packing volume (m^3)	45	10 000[a]	20 000	285
unit cost ($1000)[b]	125	4000	15 000	1770
packing material	Brentwood CF1900	Brentwood XF12560	Brentwood XF12560	Sulzer 250X
orientation	counter-flow	cross-flow	cross-flow	counter-flow
inlet area cost ($ per m^2 inlet)	4150	2300	3700	15 800[c]

[a]Standard quotes for large-scale cooling towers commonly include this volume of 'splash fill' or a lesser volume of structured packing ('film fill'), but can easily be ordered with this full volume of film fill.

[b]The unit cost values represent full 'overnight' costs. Note that the CE slab cost does include site preparation, while the cooling tower costs do not.

[c]Calculated from the APS report using total contactor equipment cost ($290 million) less packing cost ($160 million), multiplied by their 4.5 scale factor to get installed cost and divided by their 37 000 m^2 inlet area.

CE is continuing with design optimization efforts to further reduce costs, including the exploration of labour cost savings associated with shop fabrication of individual 5×5 m contactor units for subsequent field assembly. CE also has a joint development agreement with a leading cooling tower manufacturer to further examine engineering, construction and materials choices that could further reduce overall contactor structure cost.

The CE contactor design is closely related to industrial cooling towers. Table 4 compares the results from the SolTech/CE design study with vendor prices from cooling tower suppliers obtained in 2010, which provide an independent check on our cost estimates. The first system is from a manufacturer of small, modular counter-flow cooling units, and the second is from a leading manufacturer of large field-erected, cross-flow cooling towers, showing that these fully mature designs with years of optimization and refinement have managed to achieve inlet area costs of $2300 m^{-2}. We also compare against a traditional, enclosed cylindrical counter-flow packed tower, such as those commonly used in the chemical processing industry, and here we have calculated the cost and characteristics from the packed tower design used in the APS report [2, p. 37].

We claim that the designers of slab air-contactors could emulate the design characteristics and construction methods of these large-scale cooling towers to eventually achieve similarly low structural costs. The inlet area cost for CE's slab contactor falls in between the costs for comparable systems that ingest large quantities of atmospheric air, namely the small-scale specialized cooling units, and large-scale field-erected cooling towers, increasing our confidence in the results from SolTech's design study for CE's slab contactor.

The working fluid of CE's slab contactor is indeed liquid hydroxide, rather than water, but this difference was handled with minimal cost penalty by the use of affordable, hydroxide-resistant, polyvinyl chloride (PVC) packing products and liquid handling components. In fact, one key benefit of the hydroxide working fluid, which will be discussed later, is that it inhibits biological growth and the resultant fouling problems that are a constant challenge in water-based cooling tower systems.

Further, the obvious outlier in this inlet area cost comparison was the traditional packed tower. Although this packed tower design represents the lowest technical risk scenario, because they are already used for CO_2 stripping at industrial scale, the fully enclosed structure leads to a prohibitively high cost and renders this design impracticable for large-scale processing of ambient air. The technical risk trade-offs associated with moving away from the packed tower design towards the more cost-effective, open, cross-flow configuration as embodied by CE's slab contactor are further discussed in §2*g*.

(*c*) Fan and pumping energy requirements

The fans incorporated in CE's air-contactor design are commonly used in forced-draught cooling towers. These two applications are very similar in terms of required volumetric air flow rate, air velocity and duration of run-time. CE has selected a Swifter CTX series fan for the air-contactor, which will operate at an efficiency of approximately 66 per cent while moving air through the air-contactor at approximately $2 \, \mathrm{m \, s^{-1}}$. There are in fact a number of commercially available choices for the fans that would perform well in the CE air-contactor because the expected pressure drop, desired air velocity and volume of air processed are very similar to forced-draught cooling towers.

The structural study performed by SolTech included a survey of pumps and piping to determine the necessary pumps and piping diameters for the air-contactor based on the estimated liquid flows. Their analysis showed that, owing to the intermittent wetting, the liquid flows were so low that the cost of pumping equipment became insignificant, allowing CE relative freedom in choosing pumping equipment and vendors.

In a typical contactor operating condition, the energy used for the fluid pumps is approximately 15 per cent of that required for the air fans. For this simplified analysis, we simply decrease the fan efficiency to 56 per cent from 66 per cent to account for this.

(*d*) Structured packing mass-transfer media

At the core of CE's air-contactor design is a structured packing material that distributes the CO_2-absorbing hydroxide solution over a large surface area, and creates efficient contact with the air by minimizing frictional air resistance in the flow channels. The large liquid surface area is essential in order to absorb a large fraction of CO_2 from the air, and the low pressure drop is needed to minimize the fan energy requirements at high air throughputs. As we explain in this section, the material of packing construction and the flow geometry are both crucial parameters for consideration in a large-scale air-contactor design.

Table 5. Structured packing specifications.

parameter	Brentwood XF12560	Sulzer 250X
SSA (m^2 m^{-3})	210	250
weight (kg m^{-3})	40	200
maximum temperature (°C)	60	900[a]
$\Delta P/D$ (at $V_{air} = 2\,\mathrm{m\,s^{-1}}$) (Pa m^{-1})	46	75
cost ($ m^{-2})	1	6
chemical compatibility	19 M NaOH	19 M NaOH
material	PVC	316SS
orientation	cross-flow	counter-flow
compatible demisters	Brentwood XF150Max	Sulzer Mellachevron

[a]High-temperature tolerance is required for many common unit operations.

Traditional packed towers in chemical processing industries commonly use structured packing products made from stainless steel, which often cost of the order of $5 m^{-2}. Note that, while packing products are priced per volume, we find it convenient to express cost per unit surface area to allow comparison between products with different specific surface areas (SSAs; see table 5 for a comparison between two packing products). For example, at $1500 m^{-3}, Sulzer 250X costs $6 m^{-2}. On the criteria of cost-effectiveness, chemical compatibility with hydroxide, high surface area, long lifetime, debris management and low pressure drop, we have identified a more appropriate family of PVC-based packing products from the cooling tower industry, upon which to base our slab contactor design. PVC structured packing products have average costs of less than $1 m^{-2}, and so the switch from stainless steel to PVC material produces marked cost savings for a large-scale slab contactor. Moreover, plastic packings can be conditioned to offer the same NaOH wetting as steel packings, and as we describe in §3b,c, the performance of plastic tower packing exceeds that of steel tower packing in pressure drop per unit of surface area, arguably the single most important efficiency metric.

Experiments at both laboratory scale and with a multi-metre tower have been conducted by CE to quantify the CO$_2$-absorption performance of both PVC and steel packing products. Principally, in 2008, a 3 m tall packed tower was operated with steel packing in a counter-flow mode and with liquid hydroxide, to measure the CO$_2$-absorption performance and fan energy requirements. In 2010, laboratory-scale tests were conducted with a cross-flow contacting apparatus on several commercial packing products, made from both steel and PVC. During this time, CE evaluated the liquid 'hold-up' (quantity of liquid retained by a wetted packing) properties, frictional resistance to air flow (measured as pressure drop) and CO$_2$-uptake performance of these products, all under a full range of capture solution concentrations, air velocities, temperatures, relative humidity values and run-times (CE's experimental results are summarized in table 6). Once natural surface conditioning occurs, as it does in cooling towers, CE has shown that PVC packing products are able to achieve the same CO$_2$-absorption and pressure-drop performance metrics as their comparable steel counterparts. We note here that our recent study presented at GHGT-10 has shown equivalent performance

Table 6. CO_2 absorption by strong hydroxide, literature values.

source	$K_L\varepsilon$ (mm s^{-1})[a]	conditions	comments
Spector & Dodge [7]	1.6	inverted from run 85F (1 inch Berl saddles), using SSA $= 250\,m^2\,m^{-3}$, $D = 3\,m$, $V = 0.66\,m\,s^{-1}$, CF $= 0.84$	experimental
Astarita [8]	1.8[b]	converted from other units. NaOH. Conditions unclear	converted from a flux value attributed to Astarita, reported in Zeman [9]
Herzog [10]	0.1[b]	converted from other units. 1 M Ca(OH)$_2$ exposed stagnant pool	theoretical
Baciocchi *et al.* [11]	1.2	inverted from table A1, 2 M NaOH scenario, using SSA $= 250\,m^2\,m^{-3}$, $D = 10.3\,m$, $V = 4.3\,m\,s^{-1}$, CF $= 0.5$	important because this study formed the basis of the APS report
Stolaroff *et al.* [12]	2.5[b]	2.5 M NaOH	theoretical
Mahmoudkhani *et al.* [13]	2.3	inverted using SSA $= 250\,m^2\,m^{-3}$, $D = 2.6\,m$, $V = 0.8\,m\,s^{-1}$, CF $= 0.85$	experimental
Zeman [14]	1.2	converted from other units. Random packing, SSA $= 300\,m^2\,m^{-3}$. 1 M NaOH, $V =$ unreported. ε accounted for by author	experimental
APS DAC report [2]	2.0	inverted from the APS DAC tower design (APS DAC report [2], table 2.3, p. 35), using SSA $= 250\,m^2\,m^{-3}$, $D = 2.8\,m$, $V = 2.0\,m\,s^{-1}$, CF $= 0.5$	theoretical. Note discrepancy with Baciocchi *et al.* [11]

(*Continued.*)

Table 6. (*Continued.*)

source	$K_L\varepsilon$ (mm s^{-1})[a]	conditions	comments
CE[c]	2.1	inverted from empirical laboratory data. XF12560 packing with SSA $= 210\,\mathrm{m^2\,m^{-3}}$, cross-flow, 2 M NaOH, $D = 0.6\,\mathrm{m}$, $V = 1.0\,\mathrm{m\,s^{-1}}$, CF $= 0.23$	CE laboratory-scale contactor
	2.4	inverted from empirical laboratory data. Sulzer 500Y packing with SSA $= 500\,\mathrm{m^2\,m^{-3}}$, cross-flow, 2 M KOH, $D = 0.6\,\mathrm{m}$, $V = 1\,\mathrm{m\,s^{-1}}$, CF $= 0.49$	CE laboratory-scale contactor

[a]Unless indicated otherwise, we have used reported experimental conditions to invert the exponential decay in equation (2.1) to give $K_L\varepsilon$ values for this table. We identify which parameters are used to invert equation (2.1) in each case.

[b]Reported as K_L only, not $K_L\varepsilon$.

[c]CE used an in-house laboratory-scale cross-flow contacting apparatus to test Sulzer 250X and 500Y, and Brentwood XF12560, XF75, CF1200 and CF1900 packing products. Tests were conducted with CO$_2$ inlet concentrations ranging from 100 to 700 ppm, relative humidity 20–100%, temperature 20–33°C, with air velocity and solution chemistries as defined above. Only two representative measurements, for 2 M NaOH and 2 M KOH, are reported here.

in CO$_2$ absorption for two PVC and steel packing products with comparable flow geometries [15]. The long-term performance of PVC packing products with strong hydroxide solutions will be evaluated by a prototype contactor being built by CE for long-duration operation in summer 2011 to quantify key risks as detailed in §2*g*.

CE's experimental evaluation shows that commercial cross-flow packing products—PVC or steel—do not have optimized geometries for AC. Both the diffusivity of CO$_2$ in ambient air and the mass transfer properties of strong hydroxide differ from the conditions normally present in heat-transfer cooling towers or packed scrubber towers; so there is room for significant gains in contacting efficiency with specialized packing design modifications. CE is pursuing this development path with suppliers that regularly produce custom packing designs, but we constrain our discussion in this study to existing commercial products.

For the purpose of this study, we base our contactor cost analysis on Brentwood Industries XF12560 structured packing, which is a commercially available product specifically designed for use in large cross-flow cooling tower applications, similar to that of the CE slab air-contactor. XF12560 is constructed from PVC that is

completely resistant to the strong hydroxide, has an efficient cross-flow geometry that produces low air-side pressure drop and possesses a similar surface area per volume as the common stainless-steel tower packing Sulzer 250X. The fully installed cost of XF12560 PVC packing, which includes labour for installation and associated peripheral products such as supports, mounts, drift eliminators and solution re-distributors, is $250 m^{-3}. For a comparison between the characteristics of Brentwood XF12560 PVC packing product, and the default stainless-steel product Sulzer 250X, we refer the reader to CE's GHGT-10 paper [15]. In our cost analysis, we have used Brentwood specifications for pressure drop, given as equation (2.2), and a 'wetting efficiency' of 0.8, which represents the fraction of the total geometrical area on which an effective hydroxide film forms.

CE has developed a procedure of supplying liquid flow to the packing media at alternately high and low rates. Brief periods of high liquid flow rates are used to fully flush the entire packing surface, and subsequently long periods of low 'trickling flow' are used to conserve pumping energy while allowing the hydroxide solution to partly react away while capturing CO_2. Through this method, CE's slab contactor is able to minimize the pressure drop of the packing material by avoiding air-channel constriction with high liquid flows, and to minimize pumping energy requirements. In fact, pumping energy requirements are calculated to be low enough when compared with fan energy usage that they are ignored in our cost optimization procedure. A discussion of packing-associated risks, including surface wettability, droplet loss by drift and physical fouling from airborne debris, is included in §2*g*.

(*e*) Carbon dioxide absorption by strong aqueous hydroxide

The feasibility of AC using aqueous absorbers rests on the CO_2 flux into the capture solution. This metric dictates the amount of liquid surface area (in practice, achieved by liquid sprays or structured packing) required behind each unit of air intake area to absorb a given fraction of ambient CO_2 and thus drives the overall scale and financial cost of an air-contactor. We illustrate in this section that, once normalized to an overall mass transfer coefficient (we use K_L, with units of m s^{-1}), literature spanning from the 1940s to the present time reports a consistent range of experimental and theoretical values for the CO_2 mass transfer performance achievable by strong hydroxide solutions (table 6). The mass transfer coefficient is crucial to the economics of AC. If K_L was an order of magnitude lower (0.1 mm s^{-1}), AC via the wet scrubbing method would probably be infeasible, whereas if it were an order of magnitude higher (10 mm s^{-1}), an AC contactor would be relatively inexpensive.

Because the flux of CO_2 into a liquid hydroxide solution under conditions relevant to AC is proportional to the mixing ratio of CO_2 in the overlying gas, the mass transfer coefficient therefore has the units of velocity. We denote this as K_L, so that $F = \rho_{CO_2} \times K_L$, where F is the mass flux into the liquid and ρ_{CO_2} is the mass of CO_2 per unit volume of air at the surface [5].

This capture of CO_2 from bulk air via liquid hydroxide occurs in several steps: (i) air-side transport of CO_2 to the surface of the liquid film, (ii) equilibration with the liquid surface, (iii) CO_2 diffusion inwards through the film, (iv) reaction with $OH_{(aq)}^-$ to remove CO_2, and (v) replenishment via diffusion of $OH_{(aq)}^-$ from within

the bulk fluid and inward diffusion of $CO_{3(aq)}^{2-}$. For NaOH, the overall reaction is

$$2NaOH_{(aq)} + CO_{2(aq)} \rightarrow Na_2CO_{3(aq)} + H_2O_{(l)}.$$
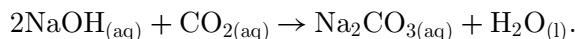
It has been shown by others [12] that a combination of the CO_2 diffusion and reaction steps (iii and iv) form the limitation to liquid-phase mass transfer, and that a reaction–diffusion equation [8,16] can be used to model this process:

$$\frac{\partial C}{\partial t} = D\nabla^2 C - \frac{1}{\tau}C. \tag{2.7}$$

Here, C denotes the concentration of CO_2 in the liquid film (in $g\,m^{-3}$), D is the diffusivity coefficient for CO_2 in the hydroxide solution (in $m^2\,s^{-1}$) and τ is the characteristic time constant (in seconds) for the rate-limiting reaction

$$CO_{2(aq)} + OH_{(aq)}^- \rightarrow HCO_{3(aq)}^-.$$
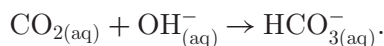
To calculate our K_L, we assume that the film of NaOH is stagnant, that the concentration profile of CO_2 in solution is a function of depth only and that the concentration profile has reached steady state. With these assumptions, we find

$$0 = D\frac{d^2 C}{dx^2} - \frac{1}{\tau}C. \tag{2.8}$$

The simplified reaction–diffusion equation is solved by using as a boundary condition the assumption that the liquid-side CO_2 concentration at the interface is $k_H C_0$, where k_H is Henry's constant for CO_2 in a hydroxide solution, and where C_0 is the air-side CO_2 concentration. The solution yields a characteristic decay length for CO_2 in hydroxide solution (equation (2.9)) and the absorption flux. The flux expression is normalized with respect to air-side CO_2 concentration, to yield the mass transfer coefficient given in equation (2.10). In these equations, k is the reaction rate constant for the limiting reaction described earlier (in $m^3\,mol^{-1}\,s^{-1}$), and $\{OH^-\}$ is the activity-corrected concentration of hydroxide [12]. Thus we have

$$L = \sqrt{\frac{D}{k\{OH^-\}}} \tag{2.9}$$

and

$$K_L = \frac{F}{C_0} = k_H\sqrt{Dk\{OH^-\}}. \tag{2.10}$$

These equations can be evaluated, for a 2 M NaOH solution as an example, using standard literature values for the parameters. Using values from Danckwerts *et al.* [17] for diffusivity $(1.21 \times 10^{-9}\,m^2\,s^{-1})$ and Henry's constant (0.64) for CO_2 in hydroxide, a reaction rate from Stolaroff *et al.* [12] for the limiting reaction $(8500\,l\,mol^{-1}\,s^{-1})$ and a hydroxide activity at 2 M $[OH^-]$ (0.66) from Marcos-Arroyo *et al.* [18], we obtain a characteristic decay length of $0.3\,\mu m$, and a mass transfer coefficient of $2.4\,mm\,s^{-1}$. At an atmospheric CO_2 concentration of 400 ppmv $(0.017\,mol\,m^{-3})$, the flux into the liquid would be $1.8\,mg\text{-}CO_2\,m^{-2}\,s^{-1}$.

Our work to characterize the CO_2-absorption performance of strong hydroxide solutions builds on theoretical and empirical work stretching back to the 1940s. Table 6 shows the conditions under which previous studies have calculated or

measured CO_2 absorption. We report $K_L\varepsilon$, which is the mass transfer coefficient multiplied by the packing wetting efficiency. The product of these coefficients determines empirically measured CO_2 absorption. We focus on the product because it is difficult to measure K_L and ε independently in a large-scale sample of packing. We obtained the $K_L\varepsilon$ values reported here by inverting the exponential decay term from equation (2.1), in the form $1 - CF = e^{-SSA D K_L\varepsilon/V}$, where CF denotes capture fraction and is defined by $CF = 1 - [CO_2]_{out}/[CO_2]_{in}$. In our cost optimization, we use a wetting efficiency of $\varepsilon = 0.8$, but other experiments, configurations or packing choices can produce values lower than this.

We note that all but one of the previously reported values, spanning a multitude of experimental conditions and theoretical parameters over many decades, are within a factor of three of one another. Given the differences in research methods used, choices in packing and apparatus, it is remarkable that these values are so closely aligned. Herzog [10] is a notable and important outlier in his choice of liquid solution, and thus mass transfer coefficient, used to evaluate the feasibility of AC. Herzog used a $Ca(OH)_2$ solution, resulting in a mass transfer coefficient that is an order of magnitude lower than those reported by studies using Na- or K-based systems with packed absorption apparatus. This discrepancy is relevant because this study and its author have been widely cited in critiques of the feasibility of AC.

For strong hydroxide solutions, in the 1–4 M range that is most applicable to AC, all of the coefficients in equation (2.10) vary strongly with concentration and speciation. While the variations in Henry's constant, diffusivity, reaction rate and activity with hydroxide concentration can be estimated from the literature, we find that the resulting predictions of K_L using equation (2.10) are typically inaccurate by factors of order unity. For this reason, we developed a set of experiments that produced an extensive database of measured mass transfer coefficients, as a function of various solution concentrations and temperatures. Our experiments were carried out under an extensive range of humidity values, inlet CO_2 concentrations, air velocities, packing types and orientation, and solution flow rates. We use this database in combination with the literature estimates for the variations of the coefficients in equation (2.10) to produce an empirical function for predicting K_L for contactor engineering.

For our baseline cost optimization scenario presented here, we choose a conservative value of $K_L = 1.5\,\mathrm{mm\,s^{-1}}$ that is representative of a partially loaded 1–2 M NaOH solution, and apply a wetting efficiency of $\varepsilon = 0.8$, to give $K_L\varepsilon = 1.2\,\mathrm{mm\,s^{-1}}$. This is lower than our own measured values by a factor of roughly 50 per cent. It is consistent with the value derived from Baciocchi *et al.* [11] and is 40 per cent lower than the value used by the APS committee [2] as derived from the report in table 6.

(f) Optimization scenarios and results

Armed with the parameters and values developed in previous sections, we now discuss the results of our cost optimization model that was itself described in §2a. We use this simple cost optimization model as a tool to direct our research efforts towards the factors that offer the most significant potential cost reductions. To illustrate the impact of parameter uncertainty, we choose three simple scenarios: 'base case' represents our current state of contactor design, 'pessimistic' gives

Table 7. Cost optimization scenarios and results.

parameter[a]	base case	pessimistic	optimistic
K_L (mm s^{-1})	1.5	0.8	2.0
C_A (\$ per m^2 inlet)	3700	5000	2500
minimum cost (\$ per tonne CO$_2$)	60	95	43
depth at minimum (m)	8.6	14.0	6.2
V_{air} at minimum (m s^{-1})	1.6	1.5	1.6

[a]All other parameters taken at baseline values from §2a.

a conservative upper bound on the cost of atmospheric CO$_2$ contacting, and 'optimistic' represents positive outcomes of current improvements under way at CE. Table 7 shows the scenarios and the resulting capture costs.

In each case, the scenario parameters were substituted into equations (2.1)–(2.5), and the resultant CO$_2$ contacting cost (equation (2.6)) was developed as a function of contactor depth D, and air velocity V. This function was then minimized over the domain of depth and velocity to find the optimal values of these variables that produced the minimal total capture cost. In our base case scenario, the minimized total cost of contacting CO$_2$ from atmospheric air was calculated as \$60 per tonne CO$_2$, and was produced with a contactor depth of 8.6 m run at an average air velocity of 1.6 m s^{-1}. Note that the CO$_2$ capture fraction (approx. 80% in our base case) is not specified; rather, it is a free design parameter value that results from the physical contactor characteristics that produce the minimum total cost. This is one important difference between AC, which extracts CO$_2$ from an essentially infinite atmospheric source and seeks only to do so at minimal cost, and traditional CCS technologies that operate on fixed gas streams. In this case, the resulting capture fractions are comparable to CCS facilities, although the design trade-offs that drive the result are different.

Our cost optimization model also calculated the fractional partial derivatives of the minimum cost with respect to the most important parameters. In our base case scenario, the fractional partial derivative of minimum cost with respect to inlet area cost was 0.54. Thus, with all other parameters held constant, a 10 per cent reduction in inlet area cost would produce a 5.4 per cent reduction in total capture cost. The fractional partial derivative for mass transfer coefficient was −0.44; for packing cost, it was 0.27; and for electricity cost, it was 0.19. Guided by these results, CE is primarily focused on reducing inlet area cost and increasing mass transfer coefficient to further reduce the total capture cost of our slab contactor design.

From table 7, it can be seen that our pessimistic scenario produces an upper bound on total contacting cost of \$95 per tonne CO$_2$, and our optimistic scenario results in a cost of \$43 per tonne CO$_2$. These two scenarios also illustrate the underlying design trade-offs calculated by our model. In the pessimistic scenario, with very large capital expense incurred by a \$5000 m^{-2} inlet contactor and the poor absorptive quality of a 0.8 mm s^{-1} solution, the optimal operating regime shifts to a very long contactor depth in order to achieve a high fractional capture of ingested CO$_2$, which in turn minimizes the contribution of the capital

expense to each tonne captured. Alternatively, with a cheap optimistic contactor running on a highly absorptive $2.0 \, \text{mm s}^{-1}$ solution, only $6.2 \, \text{m}$ of depth are needed to capture enough of the ingested CO_2 to minimize overall cost.

Optimization is crucial to producing a cost-effective contactor design. The design parameters for AC are sufficiently different from the parameters typical of CCS and of many other industrial gas separation technologies, that adopting design standards from these technologies without optimization will yield a grossly sub-optimal air-capture design.

(g) *Risks, uncertainties and operational considerations*

We consider risks in two broad categories: first, environmental health and safety risks, and second, operational risks and performance uncertainties. Risks to operators include spills or leaks in piping, but given the century of experience handling such solutions in industrial settings, we are confident that they can be managed effectively. Risks outside the plant boundary are due to dispersal of fine droplets and the presence of low-CO_2 air. These NaOH solutions are not volatile at ambient conditions; so there are minimal risks from vapour-phase transport.

Loss of hydroxide in liquid droplets is the most significant environmental risk. CE's design aims to reduce loss by two methods. First, and most importantly, the contactor will include a demister section at each face. Demisters are commercially available, easily integrated with the packing products we are working with and have acceptably low pressure drops. For example, a stock demister product XF80MAx produced by Brentwood Industries assures drift losses below 0.0005 per cent of the full fluid recirculation rate, even under the high liquid flow conditions used in cooling towers as specification. Second, our contactors operate in a low-flow regime in which there is much less energy for generating small droplets. During short periods where high-flow operation is needed for debris removal or film establishment, we have the flexibility to reduce air velocity as needed to control the particle loss rate. We expect that actual overall losses will be substantially smaller than from standard cooling towers. (Note that this must be done with care because demisters lose efficiency below some air velocity threshold.)

The central operational risk relates to the long-duration performance of packing under the low average flow conditions. Risks include fouling with airborne debris, and, in particular, some interaction of airborne contaminants and packing surface that might make it difficult to maintain wetting and thus CO_2-absorption performance.

Most liquid–gas contactors used in the chemical process industry are in the form of closed cylindrical towers operated in a counter-flow configuration with feed gas interconnections achieved via ducting that couples the gas to other process units. Let us call this a 'closed' contactor configuration. These systems are very different from 'open' configurations used to bring air into contact with an aqueous medium, important examples of which include forced-draught cooling towers, natural-draught cooling towers, and various sewage and wastewater treatment applications in which atmospheric air is brought into contact with liquid and micro-organisms growing in 'biofilms' are used to treat wastewater. Traditionally, such open applications used low-technology packing products such

Table 8. Air-contactor design risks and uncertainties.

risk	closed versus open comparison
fouling	advantage: open. Suppliers of open systems have experience managing fouling by wind-blown materials; closed systems might require pre-filters, and suppliers may lack design experience and mechanisms for managing fouling.
drift losses	advantage: closed. Easier to control drift in closed system; however open systems now have high-performance drift eliminators.
evaporative losses	advantage: unclear. Use of strong inorganic base as reagent essentially eliminates this risk for either system.
materials compatibility	advantage: closed. Designers of chemical separations towers have extensive experience with a range of corrosives, cooling towers do use alkalis but not as strong as we require.
efficiency of packing	advantage: unclear. Larger selection of high-performance packing available for closed systems, however, cross-flow cooling-tower packings have performance that exceeds common counter-flow tower packing (figure 2)
intermittent flow	advantage: unclear. Intermittent liquid flow is needed to minimizing liquid pumping work. Neither open nor closed contactors nor their packings have been designed for this.
air-side efficiency	advantage: open. Open systems designed to move large volumes of atmospheric air with low-pressure drops with little re-circulation.
cost	advantage: open. Costs are much lower.

as 'splash fill' media for cooling towers and random (e.g. crushed stone) packing for trickle filters. Over the past few decades, there has been a very strong trend in each of these applications towards use of high-performance plastic-structured packing products that cost-effectively achieve improved performance.

A useful way to assess the performance risks in the contactor design proposed here is to consider how the risk might differ for an air-capture facility based on the closed-system model when compared with one based on technologies derived from one of the open systems mentioned already. Table 8 provides comparison between the two across a number of key performance risks. Our view is that—ignoring cost and considering technology and performance risk alone—there are significant advantages to closed systems, but that there are equally or more important advantages associated with open systems because these systems have previously been designed and used to process large quantities of atmospheric air.

3. Comparison with the American Physical Society DAC report

The APS report [2] on direct air capture proposed a reference design for a contactor capturing CO$_2$ out of air using a strong (2 M) NaOH solution with an estimated overall cost for the *contactor alone* of $240 per tonne CO$_2$. The estimated costs presented here are roughly a factor of four smaller. In this section, we examine the reasons for this discrepancy. We find that the cost discrepancy is driven by design choice rather than by costing methodology. The APS report used a reference design based on closed counter-flow gas scrubber

column technology—an expensive design choice, and one that makes recirculation of outflow gas difficult to manage in a large system with multiple towers. The APS report suggested that a design such as ours, derived from a cooling tower design basis, could be significantly cheaper, but that design was ruled out of their analysis on the basis of technical risk.

(a) Cost analysis methodology

An obvious possibility for the contacting cost discrepancy between our analysis and that of the APS is that we have made unrealistic economic assumptions in scaling components to complete systems. Many academic studies of energy technologies, for example, use unrealistically low capital recovery factors. However, the APS DAC report and CE use similar costing *methods*, to scale major equipment costs up to full facility cost estimates. As described in §2*b*, our contactor cost estimate used methods consistent with the recommendations provided in the APS report, appendix A2 [2, p. 43]. In fact, CE uses *more conservative* cost assumptions. For example, both CE and APS use a 25 per cent contingency, CE assumes a 5 per cent O&M rate, whereas APS assumes approximately 2 per cent, and, while they are broken down differently, the overall capital change factor used by APS is 12 per cent while CE's is 15 per cent. The APS estimated capital costs by multiplying the raw packing cost by a scaling factor of 4.5, whereas CE's costs are based on first-order engineering studies performed by engineering procurement and construction (EPC) firms under contract to CE and on vendor quotes from a major cooling tower manufacturer with which CE has an exploratory joint-development agreement. The results are consistent in that our bottom-up SolTech study (§2*b*) did end up with a total cost that was four times the cost of packing and fans (the major equipment).

Put simply, we concur with the APS's capital cost estimate for *their reference system*, and while APS did not provide a cost for an 'open' contactor derived from cooling tower technology, they do state that such a system 'would be much less costly' (APS [2], section 2.4).

(b) Open versus closed contactor systems

The most important driver of the cost difference between the APS report and that presented here is that our design is based on an open contactor system, whereas the APS reference design is derived from closed systems. Both systems have relevance to AC, with closed systems most commonly used to perform mass transfer on ducted, often toxic, gas streams within chemical processing facilities, and open systems being the dominant choice for ingesting large quantities of ambient air for cooling applications. As we describe in §2*g*, there are performance and development risks associated with both designs. We concur with the APS that there are risks associated with the use of cross-flow packing media in open systems with strong hydroxide solutions. We take both the performance and safety risks seriously, and concur that disciplined design and engineering are required to ensure that open air-capture contactors meet existing environmental, health and safety regulations.

The APS report [2] did not offer a technical analysis of open contacting systems, but made several references to both their potential and their technical risks, namely:

— cyclic liquid distribution to the contactor packing has potential to reduce pumping costs, but has not been demonstrated (APS, p. 41);

— the practicality of cross-flow designs depends on their ability to provide good contact between the air and the sorbent (APS, p. 42);

— cross-flow designs must minimize losses of absorbent through misting (or 'drift') or reaction with atmospheric particulates (APS, p. 42); and

— cooling tower packing products are less costly than those typically used for gas scrubbing, but have SSAs of 50–$150 \, \text{m}^2 \, \text{m}^{-3}$ and are lower performance (APS, p. 49).

We address these risks in turn, to support our suggestion that open contacting systems are more relevant for cost and technical assessments of direct AC than the closed design chosen by APS.

— Cyclic liquid distribution, whereby a strong 'pulse' of liquid flow is used to fully wet the structured packing and then allowed to slowly react away with replenishment from smaller pulses, has the potential to reduce liquid pumping costs in closed or open systems. Preliminary results have been obtained from cyclic operation in a 2 m tall, 1 m wide, 6 m deep, cross-flow contactor prototype that we operated outdoors over several months. Our results have shown that, with pulsing, we can reduce average liquid flow to roughly 10 per cent of full continuous flow required for adequate wetting, while retaining an average of 80 per cent of the CO_2 capture rate attained under full flow conditions. Summary data are available at http://www.carbonengineering.com/wp-content/uploads/2011/12/CE_ OC_Pilot_2011_Results.pdf.

— Good contact between air and sorbent is provided by the choice and depth of packing material, not by contactor configuration. As long as they are wetted properly, denser packing products (with higher SSA) will produce a sharper decay of CO_2 concentration through the absorber. This comes with the trade-off that they also produce a steeper pressure gradient, and thus the choice of packing density is best accomplished within the overall contactor cost optimization method described in this study. Figure 2 demonstrates that commercial cross-flow packings can perform more than a factor of two better than the packing chosen by the APS on the ratio of pressure drop to surface area, a crucial performance metric.

— Drift losses depend on the performance of demisters. Without adequate demisters, drift losses would be a problem in either a closed or open configuration. Published data sheets from suppliers demonstrate very similar demister performance for both cooling tower applications and chemical process towers. The reason is simple; the basic physical design constraints are precisely the same. In addition, we have demonstrated with our cross-flow prototype contactor that, through our contactor configuration and careful installation of commercial demisters, we have controlled hydroxide droplet concentration at the contactor outlet to less than 10 per cent of the OHSA indoor safety regulations. Again, we refer the reader to the web-link for a summary of preliminary results.

— We return to the choice of packing product, and its impact on design and cost in §3c.
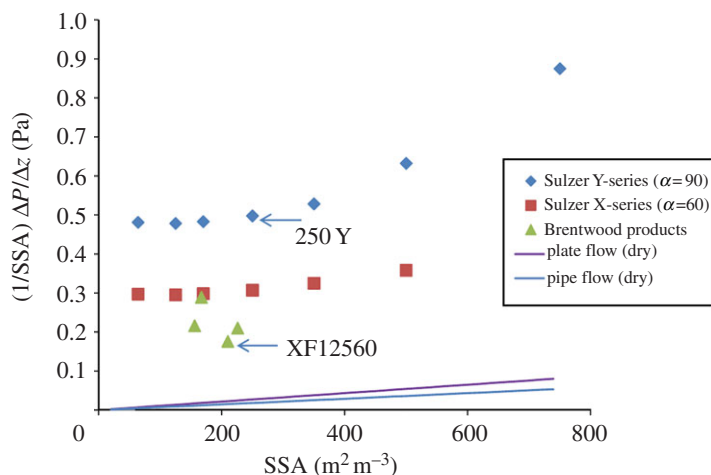
Figure 2. Pressure gradient divided by specific surface area (SSA) computed from manufacturers' data for commercial structured packing products computed at an air velocity of $2\,\mathrm{m\,s^{-1}}$ and a fluid flux of $21\,\mathrm{m^{-2}\,s^{-1}}$ ($21\,\mathrm{s^{-1}}$ supplied to each $\mathrm{m^2}$ of the top of the packing). Both CO_2 absorption rate and pressure gradient are roughly proportional to SSA; so a crucial figure of merit for the performance of structured packings is the pressure gradient divided by SSA. We have plotted the same figure of merit for flow in parallel plates and pipes as a guide to the importance of turbulent drag introduced by real-world structured packings. (Online version in colour.)

The preliminary results from CE's prototype do not resolve the risks mentioned by the APS and discussed earlier, but they do suggest that the technical risks associated with markedly more cost-effective open contactor designs can be readily resolved by inexpensive testing.

(c) Packing choice

The APS report [2] states that packing products used for cooling towers are available with SSAs between 50 and $150\,\mathrm{m^2\,m^{-3}}$ but that they have lower performance than those used for chemical process towers; the report does not evaluate their use for direct AC. This choice is important, because the purchase cost of packing was one-third of the total major equipment cost for the entire APS reference design (APS, pp. 37 and 39); yet plastic packing products similar to the 250Y model used by the APS report are commercially available at one-sixth the cost. Current suppliers of cooling tower packing products do make media with SSAs exceeding $200\,\mathrm{m^2\,m^{-3}}$, and results presented here and elsewhere demonstrate that these plastic products can be operated with identical CO_2-uptake performance to that achieved by standard scrubber tower packing under the ambient conditions relevant to direct AC. With all else held equal, a shift from the APS stainless-steel packing ($1700\,\mathrm{m^{-3}}$) to a cheaper PVC product (approx. $250\,\mathrm{m^{-3}}$) would cut $136 million of bare equipment cost from their reference design, and would translate to a reduction of $100 per tonne CO_2 in their total capture cost.

In addition to the cost reductions achieved by a switch from steel tower packing to PVC cooling tower packing, cooling tower designs also offer better performance in frictional air resistance. We evaluated specification sheet data for

two lines of Sulzer packing against several Brentwood products, on a basis of pressure drop per metre, per unit surface area provided, under the air and liquid flow rate conditions used in the APS report. Our analysis shows that, on a per surface area basis, the Brentwood packing in our design offers less than half the pressure gradient than the Sulzer 250Y used by APS. This translates directly to lower fan energy requirements, or higher throughput capacity for the contactor, and thus lower cost.

4. Discussion

We have performed our cost optimization on a slab geometry contactor that borrows characteristics from gas scrubbing towers commonly used in the chemical processing industry, and from cooling tower designs that are well suited to cost-effective bulk air processing. Because of the strong trade-offs between capital costs and operational energy requirements in air-contactor design, we claim that a first-order cost optimization is essential to achieve sensible cost estimates. We find that the cost of air contacting can be of the order of $60 per tonne CO_2 with a contactor design that results in a capture fraction of 80 per cent. This capture fraction stands in sharp contrast to some discussions in AC literature that assume values above 50 per cent to be cost-ineffective owing to the increasing marginal energy cost of capture as capture fraction increases. Our optimization, however, illustrates that the additional energy cost associated with this higher CO_2 capture fraction is warranted by the offset of amortized capital expenditure achieved by the increased capture rate.

Further, when coupled with realistic costs for the regeneration system that must complement the air-contactor to form a full direct air-capture facility, even if we use regeneration costs from the APS report itself, our cost estimates for AC are much lower than those in that report [2]. On the basis of our rough deconstruction of the APS cost analysis, the total cost for their regeneration system is $180 per tonne CO_2 captured. Coupled with our result for contactor cost of $60 per tonne CO_2 captured, and scaled to cost per tonne avoided by the same method as used by APS, this produces a total cost estimate for a full slab contactor DAC system of $343 per tonne *avoided*. (Note that, while we adopt the APS's costing methodology here for comparative purposes, we note that the APS's decision not to use on-site electricity with integrated capture increases the cost of AC for their reference system. Standard life cycle analysis methods support the view that one should count emissions associated with materials that cross the system boundary, but not with electricity generated and consumed inside it [19].)

The APS study stated that [2, p. 42]:

> *The largest uncertainty in the DAC cost is the cost of the air contactor.* It would be much less costly if an operable process could be designed with a more open system, such as is used in cooling towers. The practicality of such a system depends on the ability to provide good contact between the air and the sorbent, while minimizing physical losses of the absorbent solution through mechanisms such as misting or chemical reactions with particulates and acid gases in the air.

We have suggested that alternative contactor designs can achieve much lower costs than the packed tower reference design chosen by APS, and thus much lower overall costs for direct AC. Technical risks of open-air contactor designs

do, of course, remain to be addressed. But because contactors are inherently modular, these risks are readily studied by testing at surprisingly small scales. At Carbon Engineering, we recently ran a 1 m wide by 2 m tall by 6 m deep cross-flow contactor prototype for several months, for a total cost of order $1 million. Our preliminary data indicate that we have indeed addressed many of the key risks raised by the APS report. Given that a single full-scale 5×5 m module of CE's slab contactor could be built and piloted for under $10 million, resolution of disagreements about the costs and risks of AC will be resolved once relatively simple and high-value tests are conducted and independently reviewed.

Finally, in the context of a special issue on geoengineering, we have been asked by the editor to comment on how AC might fit in to the portfolio of technology options for managing emissions and climate risk. We see AC as a tool for emissions mitigation. Carbon Engineering is focused on using AC to enable production of transportation fuels with low carbon intensity, and, in that context, we see it as competing with alternative transportation fuels such as electricity, biofuels and hydrogen [4]. Like any large-scale industrial facility, it poses *local risks*, along with a *global benefit* in the form of low-carbon fuels, but these risks are similar in kind to risks from other energy technology, such as biofuel refineries. We anticipate that existing health, safety and environmental regulations can and should be used to regulate the risks of such technologies.

This is in sharp contrast to solar geoengineering technologies, which pose profound *global risks and benefits*, requiring new mechanisms of global governance. One of us (DWK) has argued that the costs of such technologies are sufficiently low (and the risks they pose are sufficiently high) that development should be directed only by public entities acting in the public interest. Development of solar geoengineering technology by private for-profit companies should be discouraged or, where feasible, banned outright.

The next major step in the development of AC is for companies such as Carbon Engineering to build and operate several early-stage commercial pilot projects to test the real-world costs and performance of AC. The development of such pilots under appropriate independent regulatory oversight should go a long way towards resolving uncertainties about cost and local environmental risk.

5. Summary

We have presented a conceptually simple method for the optimization of a slab geometry air-capture contactor. Design parameters for AC are sufficiently different from the parameters typical of CCS of many other industrial gas separation technologies that adopting design standards from these technologies without optimization will yield a grossly sub-optimal air-capture design. With standard industrial costing methodology applied to our slab contactor structure by an independent EPC firm, and under very similar economic assumptions as used by the APS DAC report, our optimization method produces total contactor costs of the order of $60 per tonne CO_2. Owing to the trade-off between capital and operating cost, the total cost is not simply linear in either major equipment cost or energy requirement. Finally, we note that the fourfold discrepancy between our estimate of contactor cost and that in the recent APS DAC report is due to fundamentally different design choices, insufficient optimization in the APS design and our choice of lower-cost contactor internals.

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