



Carbon dioxide capture by absorption, challenges and possibilities

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ABSTRACT

Challenges facing the use of reactive absorption as a technology for CO₂ capture on a global basis have been discussed. Special emphasis is placed on the reduction of energy requirement and environmental concerns. The relationships between fundamental properties of the absorbent system and the design, operation and performance of the absorption process are discussed on a fundamental basis, and pathways for searching new absorbent systems are identified. In particular, the use of new phase change systems is emphasized as a possibility for reducing the energy demand of these processes. Further, the need for an early investigation of the environmental properties of new absorbents is focused. Emissions of both absorbent and volatile degradation products must be below regulated norms, and methodologies for reducing these emissions to very low levels are suggested.

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1. Introduction

Absorption with chemical reaction has long been considered the most feasible route to post combustion CO₂ capture. This is due to the assumed maturity of the technology and its use in many industrial applications, e.g. acid gas removal from natural gas, CO₂ removal from reformer gases [1], but also existing smaller units producing CO₂ for nutritional purposes from exhaust gases e.g. the ABB-Lummus plant in Warrior Run, Maryland, producing about 150 ton CO₂ per day. However, the complex problem of global warming necessitates CO₂ capture on a totally different scale. About 25–30 billion tons of CO₂ are produced from anthropogenic sources annually and single point sources may be in the range of 15–20 million tons per year. Including the concept of storage of the CO₂ implies that there will be no income side to CCS unless artificially created by quota and trade mechanisms. Thus the drive for more cost and energy effective solutions for CO₂ capture is needed.

However, a global solution for CO₂ capture must also be environmentally safe. This means that we must avoid creating a new problem while solving another, no matter how serious our initial problem is. This paper will try to discuss the main challenges facing reactive absorption as a technology and what possibilities exist.

2. Energy requirement

Fig. 1 shows the main energy sinks in a conventional post combustion absorption process. For the removal of CO₂ from coal and

natural gas (NG) based power production the partial pressure of CO₂ in the exhaust gas will typically be 11–13 and 3.5–4 kPa respectively. To separate the CO₂ out and bring it up to a delivery pressure, often set to 110 bar demands energy and the lower the initial partial pressure the higher the energy demand. A decade ago the norm for a standard MEA (Monoethanolamine) based process would be a reboiler steam requirement of around 4000–4200 MJ/ton CO₂ captured for coal and NG respectively.

The development in the last years has been quite fast and the industry claims to be able to capture CO₂ from coal based exhaust with a reboiler energy requirement of about 2800 MJ/ton CO₂, or even less [3,4].

The energy used in these processes is a mix of heat to the reboiler and electricity required to drive liquid circulation pumps, but mainly for the pressure boost needed to drive the exhaust gas through the absorber including water wash, and the compressor producing the 110 bar delivery pressure. Table 1 gives an approximate distribution of the energy required for the coal and NG cases for a 400 MW power station.

The range given for the heat requirement reflects the values 4000, 4200, and 2800 MJ/ton CO₂ captured. The energy efficiencies used were respectively 42 and 58% for coal and NG. What is clear from this table is that not only do we need to address the heat requirement, but also possible reductions in booster fan and compressor energy use.

2.1. Heat requirement

As indicated in Fig. 1 the heat required for the capture goes to the reboiler but is used for three different purposes [5,6]. Firstly the exothermic reactions between CO₂ and absorbent need to be

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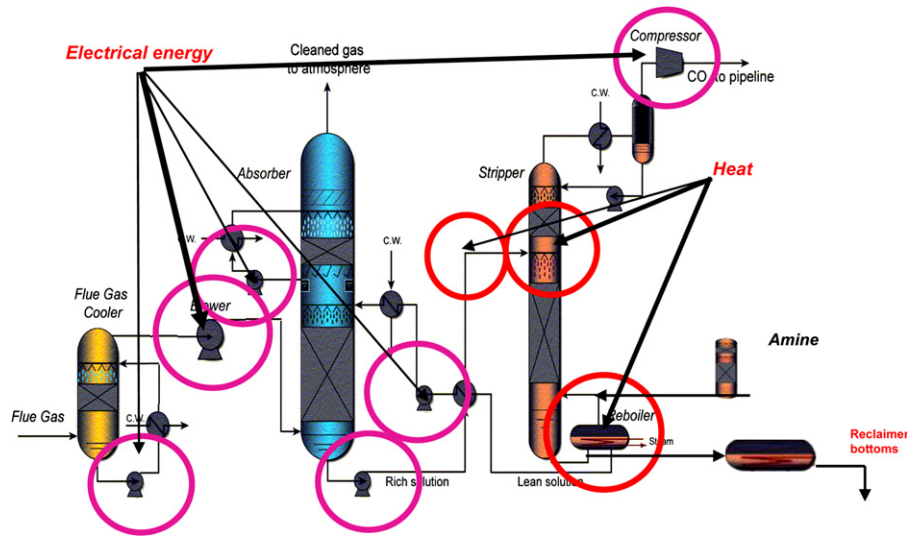


Fig. 1. Energy sinks in a conventional post combustion CO₂ capture process.

Table 1
Approximate energy requirements for post combustion CO₂ capture.

	Gas flow NM ³ /h	CO ₂ produced tons/year	Heat MW _{steam}	Heat equiv. MW _{el}	Exhaust gas boost, MW _{el}	Compression MW _{el}	Misc. MW _{el}	Prod. w/o CO ₂ capt. MW _{el}	Prod. with CO ₂ capt. MW _{el}
Coal, 12% CO ₂	1200,000	220,000	200–310	50–77.5	3.5	27	2	400	290–317.5
NG, 3.5% CO ₂	2000,000	1000,000	90–140	22.5–35	6	12	1	400	346–358.5

reversed, represented by a heat of desorption, $-\Delta H_{des}$. Secondly, to obtain a low lean amine loading, the temperature in the reboiler is normally around 120 °C. This implies a reboiler pressure of about 1.8–2 bar. The stripper has a low pressure drop so the pressure at the stripper top will also be around 1.8–2 bar. However, the CO₂ partial pressure at the top of the stripping section may well be far below this level. This implies that a steam pressure is needed to make up the total pressure and the gas mixture leaving the stripper will typically contain a high fraction of steam. This steam is condensed and returned, and the latent heat lost in the condenser. This heat required is often referred to as stripping steam. Finally, the rich amine entering the stripper cannot be brought all the way up to the reboiler temperature by the rich/lean heat exchanger, and this represents what may be called a sensible heat requirement. The sensible heat requirement is a purely process determined heat loss as long as desorption does not take place in the heat exchanger. However, the heat of desorption and the need for stripping steam are strongly interrelated and also related to the process design.

In [7] an overview of heats of absorption for different amines is given. The heat of absorption varies with both temperature and CO₂ loading, and there is a clear distinction between the different classes of amines. All the primary amines have heats of absorption around 80–90 kJ/mol CO₂, the secondary are at about 70–75 kJ/mol CO₂ and the tertiary at about 55 kJ/mol CO₂. It may thus seem tempting to go for tertiary amines since this would lower one major heat sink. However, the heat of reaction is strongly linked to both the stripping steam requirement through the absorption equilibrium temperature sensitivity, and to the rate of absorption.

The link between heat of reaction and temperature sensitivity can fundamentally be described by the Gibbs–Helmholtz equation. The temperature dependency of the Gibbs energy may be written as [8]:

$$\left(\frac{\partial G}{\partial T}\right)_p = \frac{G - H}{T} \quad (1)$$

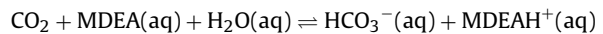
After some manipulations this may be written as [9]:

$$\left(\frac{\partial(G/T)}{\partial(1/T)}\right)_{p,n} = H \quad (2)$$

Further, since $\Delta G = -RT \ln K$, this can be substituted into Eq. (2) and we obtain the so-called van't Hoff's equation

$$\frac{\partial}{\partial(1/T)}(\ln K)_{p,n} = \frac{-\Delta H}{R} \quad (3)$$

An overall reaction for e.g. the absorption of CO₂ into aqueous MDEA may be written as



As an overall reaction this may be seen as the sum of several primary reactions. The equilibrium constant for this overall reaction may be written as:

$$K = \left(\frac{x_{\text{HCO}_3^-} \cdot x_{\text{MDEAH}^+}}{(p_{\text{CO}_2}/p^0) \cdot x_{\text{H}_2\text{O}} \cdot x_{\text{MDEA}}}\right) \cdot \left(\frac{\gamma_{\text{HCO}_3^-} \cdot \gamma_{\text{MDEAH}^+}}{\phi_{\text{CO}_2} \cdot \gamma_{\text{H}_2\text{O}} \cdot \gamma_{\text{MDEA}}}\right) \quad (4)$$

Inserted into Eq. (3) this gives

$$\left(\frac{\partial \ln K}{\partial(1/T)}\right)_{p,n} = \frac{\partial}{\partial(1/T)} \left[\ln p^0 + \ln \left(\frac{x_{\text{HCO}_3^-} \cdot x_{\text{MDEAH}^+}}{x_{\text{H}_2\text{O}} \cdot x_{\text{MDEA}}}\right) + \ln \left(\frac{\gamma_{\text{HCO}_3^-} \cdot \gamma_{\text{MDEAH}^+}}{\phi_{\text{CO}_2} \cdot \gamma_{\text{H}_2\text{O}} \cdot \gamma_{\text{MDEA}}}\right) - \ln p_{\text{CO}_2} \right] = \frac{-\Delta H}{R} \quad (5)$$

Since the standard state does not change with temperature, the derivative of the first term in the square brackets can be disregarded. If neither speciation nor the activity constant ratio changes with temperature, and the excess enthalpy is assumed to be zero, Eq. (5) reduces to

$$\frac{\partial}{\partial(1/T)}(\ln p_{\text{CO}_2})_{p,n} = \frac{-\Delta H}{R} \quad (6)$$

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