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Review

A techno-economic review on carbon capture, utilisation and storage systems for achieving a net-zero CO_2 emissions future

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ABSTRACT

Carbon capture and storage (CCS)/carbon capture, utilisation and storage (CCUS) systems are widely recognised to have the potential in reducing CO_2 emissions. However, current their global deployment is still not sufficient to reach the anticipated net-zero CO_2 emissions target by 2050. This article aims to provide a general techno-economic review of CCUS systems. The technology readiness, technical performance, energy requirement and cost associated with CO_2 capture, separation, transport, utilisation and storage technologies were discussed and compared. The CO_2 capture technological pathways include industrial separation, post-combustion, pre-combustion, oxy-fuel combustion, chemical looping combustion and direct air capture. CO_2 separation technologies such as absorption, adsorption, membrane, cryogenic and biological were also covered. Then, a review on CO_2 transportation by pipeline, ship, truck and rail was presented, followed by a review on CO_2 utilisation pathways for direct usage and through conversion into other products. Lastly, different CO_2 storage options were reviewed, which include storage through CO_2-enhanced oil recovery, in depleted oil and gas fields, in saline formations, in basalt and ultramafic rocks, in coal seams through enhanced coal bed methane recovery and in the deep ocean. This article concluded that the challenges with current CCUS technologies can possibly be overcome by developing a commercially viable hybrid system comprising more than one technology. However, this approach needs to be further investigated for industrial applications.

1. Introduction

Carbon dioxide (CO_2) is a primary anthropogenic greenhouse gas (GHG) that accounts for about 76% (38 Gigatonnes (Gt) CO_2) of the global GHG emissions in 2010 and it was contributed from the energy supply (35%), agriculture, forestry and other land use (24%), industry (21%), transport (14%) and buildings (6%) sectors (Masson-Delmotte et al., 2014). Although the global CO_2 emissions dropped by 5.8% (almost 2 Gt CO_2) in 2020 due to decreased demand for coal, oil and gas (resulted from extending restrictions on movement during the coronavirus pandemic) and increased demand for renewable energy, the global energy-related CO_2 emissions remained at 31.5 Gt (International Energy Agency (IEA), 2021a) that caused the global annual mean of atmospheric CO_2 concentration to rise by 2.6 parts per million (ppm) (i.e., 412.5 ppm, about 50% above pre-industrial levels in 1750) in 2020 (National Oceanic and Atmospheric Administration NOAA, 2021a). If the demand for coal, oil and gas rebounds with the economy, the global energy-related CO_2 emissions are projected to rebound and grow by almost 5% (1,500 Megatonnes (Mt) CO_2) in 2021, with emissions of about 1.2% (400 Mt CO_2) below 2019 emissions levels (International Energy Agency (IEA), 2021a).

The current increase in the anthropogenic emissions of atmospheric CO_2 and other GHGs causes the global annual average surface temperature of the Earth to rise by 1 °C in 2020, which was the hottest year on record since 1880 (National Oceanic and Atmospheric Administration (NOAA), 2021b). Despite worldwide efforts to reduce CO_2 and GHG emissions, the Intergovernmental Panel on Climate Change (IPCC) forecasted that the Earth's temperature will rise by 1.5 °C between 2030 and 2052 if it continues to increase at the current rate (Masson-Delmotte et al., 2019). This indicates that more actions need to be taken to limit the Earth's temperature increase to no more than 2 °C above pre-industrial levels to prevent catastrophic consequences on climate change (Fletcher and Smith, 2020). To limit global warming to below 1.5 °C, global net anthropogenic CO_2 emissions need to decline by about 45% from 2010 levels by 2030, reaching net-zero CO_2 emissions (i.e., when anthropogenic CO_2 emissions are balanced globally by anthropogenic CO_2 removals from the atmosphere) around 2050 (IPCC, 2018). For limiting global warming to below 2 °C, global net anthropogenic CO_2 emissions need to decline by about 25% by 2030 in most pathways and reach net-zero CO_2 emissions around 2070 and global GHG emissions would have to be reduced by 60% to 80% by 2050 so that the atmospheric CO_2 concentrations can stay at about or below 450 ppm by 2050 (IPCC, 2018; Hassol, 2011).

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Nomenclature

Acronyms

AR	Air reactor
ASU	Air separation unit
BECCS/BioCCS	Bioenergy with carbon capture and storage
CCS	Carbon capture and storage
CCU	Carbon capture and utilisation
CCUS	Carbon capture, utilisation and storage
CLAS	Chemical looping air separation
CLC	Chemical looping combustion
CLOU	Chemical looping with oxygen uncoupling
DAC	Direct air capture
DRI	Direct reduced iron
EBN	Energie Beher Nederland
ECBM	Enhanced coal bed methane recovery
EGR	Enhanced gas recovery
EOR	Enhanced oil recovery
ESA	Electric swing adsorption
FR	Fuel reactor
FTMs	Facilitated transport membranes
GHG	Greenhouse gas
IGCC	Integrated gasification combined cycle
IPCC	Intergovernmental Panel on Climate Change
LCOE	Levelized cost of electricity
LEILAC	Low Emissions Intensity Lime and Cement
LNG	Liquefied natural gas
LPG	Liquified petroleum gas
MMMs	Mixed matrix membranes
NGCC	Natural gas combined cycle
OTM	Oxygen-transport membrane
PE	Pore-expanded
PIM	Polymers of intrinsic microporosity
PSA	Pressure swing adsorption
SCPC	Supercritical pulverized coal
STP	Standard temperature and pressure
TRL	Technology readiness level
TSA	Temperature swing adsorption
TVSA	Temperature and vacuum swing adsorption
UK	United Kingdom
US	United States
USC	Ultra-supercritical
VPSA	Vacuum and pressure swing adsorption
VSA	Vacuum swing adsorption
VTSA	Vacuum and temperature swing adsorption
WGS	Water-gas shift
ΔH^0_{298}	Standard enthalpy of reaction
% vol.	By volume
% wt.	By weight

Chemicals

AC	Activated carbon
AHA	Aprotic Heterocyclic Anion ILs
Al_2O_3	Alumina/Aluminium oxide
AMP	2-amino-2-methyl-1-propanol
BPEI	Branched-polyethylenimine
$CaCO_3$	Calcium carbonate
CaO	Calcium oxide
$Ca(OH)_2$	Calcium hydroxide
CF_4	Carbon tetrafluoride
CH_3OH	Methanol
CH_4	Methane
CNTs	Carbon nanotubes
CNT(APTS)	CNTs modified by 3-aminopropyl-triethoxysilane
CO	Carbon monoxide
CO_2	Carbon dioxide
$CO(NH_2)_2$	Urea
Cu	Copper
Cu14–γAl	14% wt. Copper impregnated on gamma-alumina (γ–Al_2O_3)
CuBTC	Copper(II) benzene-1,3,5-tricarboxylate
DEA	Diethanolamine
DEPG	Dimethyl ether of polyethylene glycol
DETA	Diethylenetriamine
DT(NNN)	Diethylenetriaminopropyl
DMC	Dimethyl carbonate
EDA	Ethylenediamine
[emim][Tf_2N]	1-ethyl-3-methylimidazolium-bis(trifluoromethylsulfonyl) imide
FeBTC	Iron 1,3,5-benzenetricarboxylate
Fe_2O_3	Iron oxide
H_2	Hydrogen
HCl	Hydrogen chloride
H_2O	Water
H_2S	Hydrogen sulfide
[hmim][Tf_2N]	1-Hexyl-3-methyl-imidazolium bis(trifluoromethylsulfonyl)imide-based IL
HMS	Hexagonal mesoporous silica
ILs	Ionic liquids
KOH	Potassium hydroxide
Li_4SiO_4	Lithium orthosilicate
Li_2ZrO_3	Lithium zirconate
MDEA	Methyldiethanolamine
MDEA/PZ	Blend of MDEA and PZ
Me	Metal
MEA	Monoethanolamine
MeO	Metal oxide
MgO	Magnesium oxide
MOFs	Metal-organic frameworks
N_2	Nitrogen
Na_2CO_3	Sodium carbonate
NaOH	Sodium hydroxide
NH_4F	Ammonium fluoride
Ni18–αAl	18% wt. nickel impregnated on alpha-alumina (α–Al_2O_3)
NMP	N-methyl-pyrrolidone
NO_x	Nitrogen oxides
O_2	Oxygen
[P_{2228}][CNPyr]	Triethyl(octyl)phosphonium 2-cyanopyrrolide
[P_{66614}][4-ABI]	4-azabenzimidazole-based IL
[P_{66614}][2-CNPyr]	Trihexyltetradecylphosphonium 2-cyanopyrrolide
[$P_{14,6,6,6}$][Tf_2N]	Trihexyl(tetradecyl)phosphonium bis(trifluoromethylsulfonyl)imide-based IL
PC	Propylene carbonate
PEHA	Pentaethylenehexamine
PEI	Polyethylenimine
PI	Polyimide
PIP	Piperazine
PMMA	Poly(methyl methacrylate)
PS	Polysulfone
PVAm	Polyvinylamine
PVC	Polyvinyl chloride

PVDF	Polyvinylidene fluoride
PZ	Piperazine
SO_2	Sulphur dioxide
SO_x	Sulphur oxides
TEPA	Tetraethylenepentamine
TiO_2	Titanium dioxide
TRI	Triamine-containing silane
ZAPS	Erionite
ZN-19	Clinoptilolite
ZrO_2	Zirconium dioxide
ZnO	Zinc oxide
ZNT	Mordenite

Units

bbl	Barrels
Gt	Gigatonne
kW_{th}	Kilowatt-thermal
kt	Kilotonne
MJ	Megajoule
Mt	Megatonne
MW	Megawatt
MWh	Megawatt-hour
ppm	Parts per million
ppmv	Parts per million by volume
t	Tonne

There are several ways to reduce global CO_2 emissions, which include enhancing energy efficiency, increasing renewable energy production, implementing a carbon tax, planting trees, conserving existing forests and grasslands, and capturing CO_2 from power plants and other sources (Nunez, 2019). Among the CO_2 mitigation options, carbon capture and storage (CCS) is widely recognised as having the potential in meeting the world's climate change targets, allowing continued use of fossil-fuelled power plants whilst reducing CO_2 emissions into the atmosphere (Bui et al., 2018; Boot-Handford et al., 2014). CCS involves capturing or separating CO_2 from industrial and energy-related sources, transporting it to a storage site and isolating it from the atmosphere for a long period of time (Metz et al., 2005). Often, CCS is used interchangeably with the term carbon capture, utilisation and storage (CCUS), which includes the use of captured CO_2 for other applications (IEA 2021b). In response to the rising climate ambition, CCS projects around the world are growing robustly and there are 135 commercial CCS facilities in the project pipeline in 2021 (in which 27 are fully operating, 4 are under construction, 102 are under development and 2 are suspended) with an estimated total mean CO_2 capture capacity of 149.4 Mt CO_2/year in September 2021 (Turan et al., 2021). However, despite the acceleration of CCS projects, much more deployment of the technology and commitment to climate action through active market support and emissions regulation are required to reach global climate goals by 2050 (Turan et al., 2021; Scott et al., 2013).

The purpose of this article is to provide a general review of CCUS systems by assessing their technology readiness, technical performance, energy requirement and cost. First, the current status of global CCS/CCUS developments and applications in various sectors are presented in Section 2. Then, an overview of CCUS technologies is covered in Section 3, which includes the techno-economic reviews on CO_2 capture (Section 3.1), separation (Section 3.2), transport (Section 3.3), utilisation (Section 3.4) and storage (Section 3.5). Lastly, the article concludes with research prospects for CCUS systems.

2. Current status of global CCS/CCUS developments and applications

2.1. Global CCS/CCUS projects

The interest and support for CCS/CCUS are growing globally with steady progress on climate action commitments, however greater ambition and more action are required to reach 2050 climate goals (Gebremedhin et al., 2021). As seen in Table 1, the global front runner in CCS/CCUS deployment was still in the Americas region with 41 new CCS/CCUS projects announced in 2021 and this is possibly due to stronger climate commitments, the return of the United States (US) to the Paris Agreement, finalisation of Section 45Q carbon capture tax credits regulations and anticipated global demand for low carbon fuels and products (Turan et al., 2021). The CO_2 capture capacity of all commercial CCS/CCUS facilities in the Americas region was the largest compared to other regions in the world with 89% growth in total maximum CO_2 capture capacity, from 56.1 Mt CO_2/year in 2019 to 106.1 Mt CO_2/year in September 2021 (Figure 1). This was because of an increase in large-scale CCS/CCUS projects and a large proportion of facilities were in advanced development in the US, as shown in Figure 2.

Europe was the second leading region in CCS/CCUS deployment with 25 new CCS projects added in 2021 (Table 1), owing to an ambitious target to reach net-zero emissions by 2050, greater diversity of CCS projects in development in various sectors (for examples, direct air capture (DAC) and hydrogen production), increase in CCS networks for larger volumes of CO_2 storage (Department of Energy & Climate Change, 2012) and the United Kingdom (UK) Government's £1 billion CCUS infrastructure funding to establish 4 CCUS industrial clusters across the UK to capture 10 Mt CO_2/year by 2030 (Department for Business, Energy & Industrial Strategy, 2021). The increase in CCS projects in the Europe region resulted in 32% rise in the total maximum CO_2 capture capacity of all commercial CCS facilities in the region, from 28.4 Mt CO_2/year in 2020 to 37.4 Mt CO_2/year in September 2021 (Figure 1). Majority of the CCS projects in the Europe region as of September 2021 were in early development (Figure 2).

The third highest number of CCS projects as of September 2021 was seen in the Asia Pacific region with 5 new CCS projects, attributed to the inclusion of CCS in the Emissions Reduction Fund by the Australian Government that provides the first financial incentive scheme for CCS in the region (Steyn and Havercroft, 2021) and net-zero CO_2 emissions commitments by two large corporations headquartered in developed countries (Petronas in Malaysia and Repsol in Indonesia) despite insufficient policy support (Turan et al., 2021). The total maximum CO_2 capture capacity of all commercial CCS facilities in the Asia Pacific region has grown by 46%, from 10.3 Mt CO_2/year in 2020 to 15.0 Mt CO_2/year in September 2021 (Figure 1) due to the increasing number of large-scale CCS projects in the region. As of September 2021, CCS projects in the Asia Pacific region were mostly in early development (Figure 2).

The number of CCS projects in the Middle East region was the lowest compared to other regions in the world (Table 1). However, they already account for 10% of global CO_2 captured annually with a total maximum CO_2 capture capacity of 3.8 Mt CO_2/year in September 2021 from 3 operational CCS facilities in the region, which was twice of that in the Europe region (Figure 2).

2.2. Applications of CCS/CCUS technologies

CCS/CCUS projects are becoming more diverse in a broad range of applications such as natural gas processing, fertiliser production, ethanol production, chemical production, hydrogen production, power generation from coal and natural gas, waste-to-energy, iron and steel production, cement production and DAC, as shown in Figure 3. CCS/CCUS projects in power generation plants, particularly in the Americas and Europe regions, have the largest proportion of global CO_2 capture capacity compared to other sectors and it accounts for 37% of global

Table 1
Commercial CCS/CCUS projects worldwide by region and country from 2019 to September 2021. Data from the Global CCS Institute (Turan et al., 2021; Page et al., 2020; Page et al., 2019).

Region	2019		2020		September 2021	
Americas	Brazil = 1 Canada = 4 US = 19	24	Brazil = 1 Canada = 4 US = 33	38	Brazil = 1 Canada = 8 US = 70	79
Asia Pacific	Australia = 3 China = 8 South Korea = 1	12	Australia = 2 China = 6 New Zealand = 1 South Korea = 1	10	Australia = 3 China = 6 Indonesia = 2 Malaysia = 1 New Zealand = 1 South Korea = 1	14
Europe	Ireland = 1 The Netherlands = 2 Norway = 3 UK = 6	12	Ireland = 1 The Netherlands = 1 Norway = 4 UK = 7	13	Belgium = 4 Denmark = 1 Hungary = 1 Ireland = 1 Italy = 2 The Netherlands = 6 Norway = 5 Sweden = 2 UK = 16	38
Middle East	Saudi Arabia = 1 United Arab Emirates = 2	3	Qatar = 1 Saudi Arabia = 1 United Arab Emirates = 2	4	Qatar = 1 Saudi Arabia = 1 United Arab Emirates = 2	4
Total		**51**		**65**		**135**

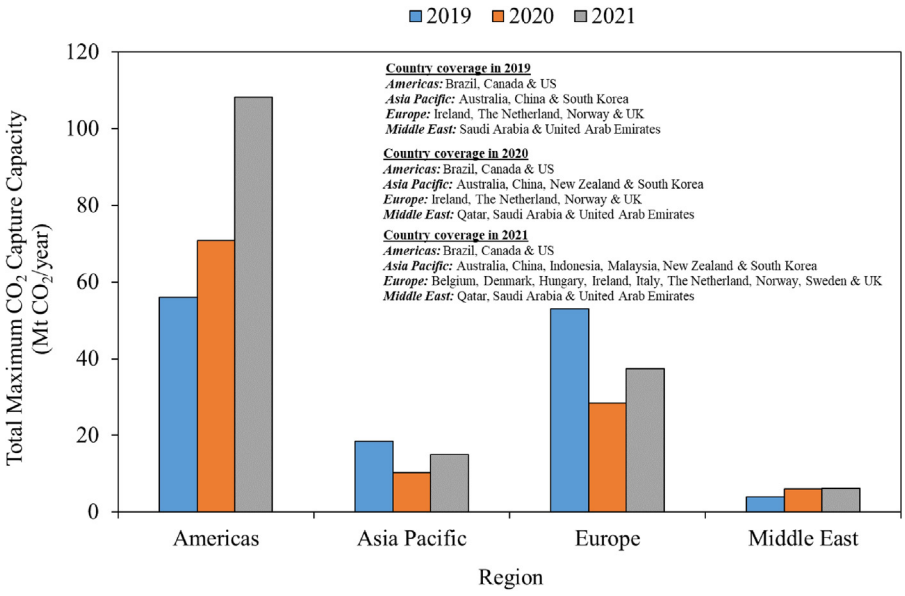

Figure 1. Total maximum CO_2 capture capacity of commercial CCS/CCUS projects worldwide from 2019 to September 2021. Data from the Global CCS Institute (Turan et al., 2021; Page et al., 2020; Page et al., 2019).

CO_2 captured annually with a total maximum CO_2 capture capacity of 62.5 Mt CO_2/year in September 2021 despite 13 projects fewer than in ethanol production plants in the US (Figure 3). The sector with the second largest global CO_2 capture capacity was the natural gas processing, mainly in the Asia Pacific and the Middle East regions, which accounts for 26% of global CO_2 captured annually with a total maximum CO_2 capture capacity of 42.6 Mt CO_2/year in September 2021 (Figure 3).

2.3. Global CCS/CCUS networks

Recently, there has been a rising trend in the development of industrial hubs with shared CO_2 transport and storage infrastructure (such as pipelines, shipping, port facilities and storage wells), a transition from large, stand-alone CCS and CCUS facilities. This approach could reduce unit costs through economies of scale, reduce commercial risk and financing costs by separating out the capture, transport and storage components, and create new investment opportunities (IEA, 2020a). For example, the Summit Carbon Solution network in the Americas region, an emerging world largest CCS project, is supporting 31 separate bioethanol plants with CO_2 transport and storage capacity of 11 Mt CO_2/year (Submit Carbon Solutions, 2021).

In the Europe region, Porthos network in Rotterdam, Netherlands, an emerging world CCUS and hydrogen hub, shares pipeline to transport CO_2 captured from refineries and 4 new blue hydrogen-producing facilities (operated by Air Products, Air Liquide, ExxonMobil and Shell) in the Port of Rotterdam area to offshore storage beneath the North Sea, about 20 km off the coast, with an initial storage capacity of 2.5 Mt CO_2/year (Porthos, 2021; Hydrocarbons Technology, 2021). Blue hydrogen (or low-carbon hydrogen) is produced from natural gas by steam reforming and the carbon emissions are captured and stored (National Grid, 2021). Another world-scale CCS hub in the Netherlands is the Aramis project (jointly developed by TotalEnergies, Shell, Energie Beher Nederland (EBN) and Gasunie) provides CO_2 transport facilities with a capacity

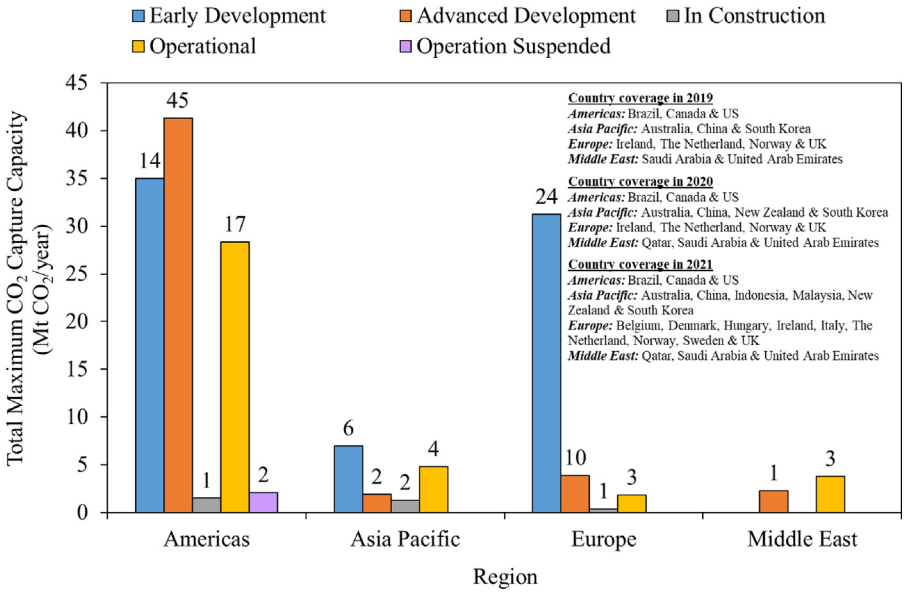

Figure 2. Total maximum CO_2 capture capacity of commercial CCS/CCUS projects worldwide at various technology development stages in September 2021. The number labelled above each column denotes the number of CCS/CCUS projects. Data from the Global CCS Institute (Turan et al., 2021).

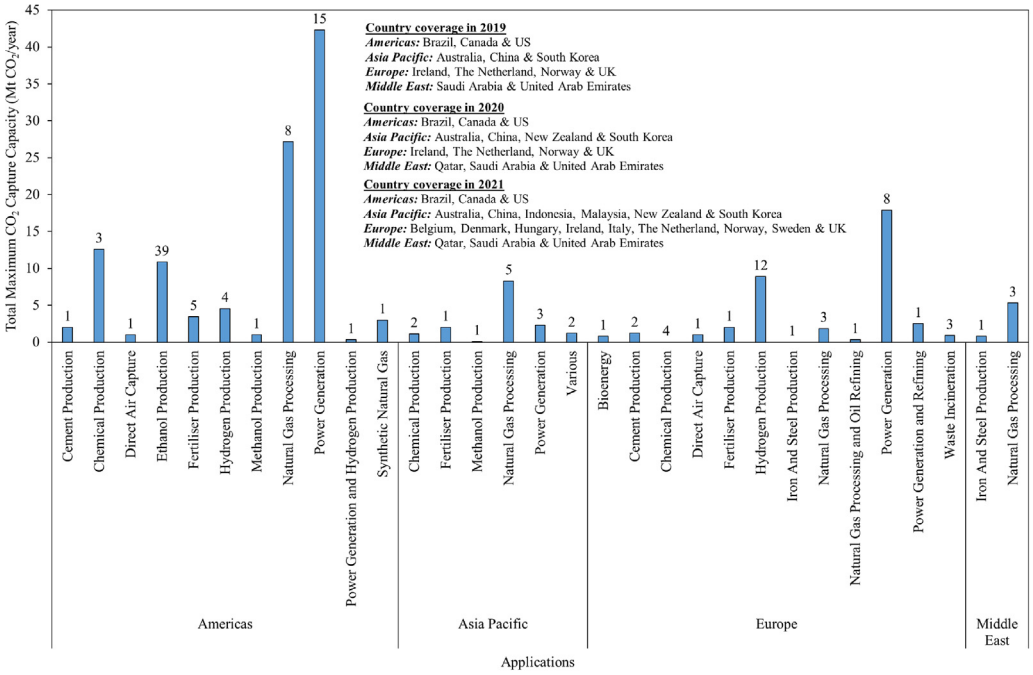

Figure 3. Total maximum CO_2 capture capacity of commercial CCS/CCUS projects worldwide by applications and regions as of September 2021. The number labelled above each column denotes the number of CCS projects. Data from the Global CCS Institute (Turan et al., 2021).

of about 5 Mt CO_2/year to offshore CO_2 storage sites (total storage capacity of more than 400 Mt CO_2), 3 to 4 km beneath the North Sea, for a variety of hard-to-abate industries (such as iron and steel cement and chemicals) (Aramis, 2021).

Moreover, the Longship (or Langskip in Norwegian) project in Norway will be the first cross-border, open access CO_2 transport and storage infrastructure network in the Europe region. CO_2 will be captured from Norcem cement plant in Brevik and Fortum Oslo Varme waste-to-energy plant in Oslo, liquified and transported by ship to an onshore terminal on the Norwegian west coast, then offloaded and pumped through a pipeline to offshore storage (about 3 km below the seabed) beneath the North Sea for permanent storage. The CO_2 transport and storage facility are managed by Northern Lights Joint Venture, developed by Equinor, Shell and Total (Northern Lights, 2021). The Longship CCS network was planned for initial storage of 1.5 Mt CO_2/year over 25 years of operation and 5 Mt CO_2/year in Phase 2 (Norwegian Ministry of Petroleum and Energy, 2021).

There has been significant development on CCS hub and cluster projects across the UK in recent years. Humber Zero in England is a ground-breaking green project that can remove up to 8 Mt CO_2/year by 2030 from the Immingham industrial side. This project integrates CCS technologies and hydrogen production within an existing industrial cluster for large-scale decarbonisation in the Humber region and the CO_2 captured was transported via pipeline either to CO_2 storage fields in the North Sea or exported to international markets from the Port of Immingham (Humber Zero, 2021). The East Coast Cluster in England is one of the UK's first CCS projects that can remove 50% of the UK's industrial cluster CO_2 emissions and it supports two of the UK's

leading industrial decarbonisation consortia (Zero Carbon Humber and Net Zero Teesside) by providing a common infrastructure to transport CO_2 from emitters across the Humber and Teesside to secure offshore storage in the Endurance aquifer in the Southern North Sea (East Coast Cluster, 2021).

The Acorn project is a low-cost, low-risk CCS project and it is a major CCS and hydrogen hub at St Fergus gas terminal in North East Scotland that reused existing onshore oil and gas pipelines to transport initially about 0.3 Mt CO_2/year captured from existing CO_2 emitters (gas processing units) at St Fergus gas terminal to offshore CO_2 storage site, about 100 km from St Fergus and over 2.5 km under the seabed (Acorn, 2021). HyNet North West is an innovative low carbon and hydrogen energy project that plans to produce, store and distribute hydrogen as well as capture and store carbon from industry in the North West of England and North Wales from 2025. This project could reduce CO_2 emissions by 10 Mt annually and deliver 80% of the UK's clean power target for transport, industry and homes by 2030 (HyNet, 2021). The South Wales Industrial Cluster project is supported by a range of partner organisations in South Wales to create the world's first net-zero emissions industrial zone by developing low carbon power, hydrogen and CCUS infrastructure for the area (Costain, 2021).

A world-class, large-scale, multi-user CCS network in the Asia Pacific region is the CarbonNet project in Victoria, Australia (in advanced development) that transport CO_2 captured from a range of industries based in Victoria's Latrobe Valley via a pipeline and injecting it deep into offshore underground storage sites in the Gippsland Basin, with an estimated initial capacity of 2 Mt CO_2/year to 5 Mt CO_2/year that can potentially increase significantly to over 20 Gt CO_2 in the future (Victorian Department of Primary Industries, 2012). Another large-scale CCS network in the Asia Pacific region is the Junggar Basin CCS hub in Xinjang, China and it has a storage capacity of 0.2 Mt CO_2/year to 3 Mt CO_2/year that is expected to increase to 10 Mt CO_2/year by 2030 (Fan, 2021). A fully operational CCS network in the Middle East region is the Abu Dhabi cluster in the United Arab Emirates that will transport 2.7 Mt CO_2/year to 5 Mt CO_2/year to nearby large onshore and offshore oil and gas fields via pipeline for enhanced oil recovery (EOR) (Turan et al., 2021; Al Keebali, 2020).

3. Overview of CCUS technologies

Despite extensive research and development for better and more cost-effective CCS/CCUS technologies, existing projects have been slow in their delivery and there were many challenges in the investment and deployment of CCS/CCUS projects globally (Martin-Roberts et al., 2021). Improvements in existing technologies and breakthrough technological innovations can reduce the costs and enable the deployment of CCS/CCUS in new applications (Kearns et al., 2021). The most expensive element of CCS/CCUS is the CO_2 capture process, which accounts for 50% of the total costs and up to 90% with compression, and the less cost-intensive elements are transportation, utilisation and storage of CO_2 (Kheirinik et al., 2021). Figure 4 shows the current development progress of some CCUS technologies in terms of technology readiness level (TRL), which has three scale-up stages: (i) research (TRLs 1 to 3), (ii) development (TRLs 4 to 6) and (iii) demonstration (TRLs 7 to 9) (IEAGHG, 2014). The technical and economic review on CO_2 capture, separation, transport, utilisation and storage are covered separately in Sections 3.1 to 3.5.

3.1. CO_2 capture

There are several technological pathways available for CO_2 capture, which are industrial separation, post-combustion, pre-combustion, oxy-fuel combustion, chemical looping combustion (CLC) and DAC systems (Figure 5). Industrial separation and post-combustion capture technologies are widely used (from TRLs 1 to 9), followed by pre-combustion (from TRLs 3 to 9), oxy-fuel combustion (from TRLs 4 to 7) and DAC (TRL 7) while CLC is still in the development stage (TRL 6) (refer to Figure 4). The advantages and disadvantages of these CO_2 capture technologies are summarised in Table 2 while their technical and economic performances are compared in Table 3. The efficiency of the CO_2 capture/removal must be reduced to make the CCS/CCUS technology more cost-effective to be implemented as the cost of CO_2 capture from flue gas produced from power plants can escalate the price of electricity production up to 90% (Kheirinik et al., 2021).

3.1.1. Industrial process separation and capture system

The industrial sector is one of the largest sources of anthropogenic CO_2 emissions. CO_2 from industrial process streams (for examples, in the purification of natural gas and production of steel, cement and ammonia) can be captured using methods similar to post-combustion, pre-combustion and oxy-fuel combustion capture systems (Metz et al., 2005). Depending on the source, raw natural gas contains different concentrations of CO_2 that must be removed before the natural gas can be sold and the captured CO_2 is normally vented to the atmosphere (Page et al., 2020). Current commercial CCUS facilities around the world are mostly associated with natural gas processing, which captures about 30 Mt CO_2/year, and they offer relatively low-cost capture opportunities (IEA, 2021c). The cost of CO_2 capture for industrial processes with high concentration CO_2 (for examples, natural gas processing, fertiliser, bioethanol and ethylene oxide production), excluding downstream CO_2 compression, in 2020 was assumed to be less than USD 10/tonne (t) of CO_2 captured for CO_2 conditioning (Kearns et al., 2021).

The iron and steelmaking industry produced 2.6 Gt CO_2 emissions in 2019 and this sector is yet to meet its decarbonisation goals as there is only 1 operating CCS plant in this sector in Abu Dhabi, United Arab Emirates and 1 under development in the Netherlands (Turan et al., 2021). Coal or natural gas are typically used as a reducing agent in the direct reduced iron (DRI) unit to transform iron ore to elemental iron for use in steelmaking and produces pure CO_2 (Page et al., 2020). An example of a new, cost-efficient and environmentally friendly ironmaking process is the COREX process, which uses lower grade (or non-coking) coal for blast furnace operation, lump iron ore or pellets and pure oxygen to produce CO_2 and water (H_2O) vapour in the top gas that must be removed so that carbon monoxide (CO) and hydrogen (H_2) can be heated to reduce iron oxide to iron in the secondary shaft kiln (Metz et al., 2005; Primetals Technologies, 2021). Some early opportunities in CO_2 capture from the iron and steel industry are: (i) retrofitting existing blast furnaces by feeding it with oxygen (O_2) and recycled CO_2-rich top gas to convert the furnace from air firing to oxy-fuel firing with CO_2 capture and (ii) the use of hydrogen derived from fossil fuel in a pre-combustion capture step as a reducing agent for the iron ore (Metz et al., 2005).

The cement industry is another large emitter of CO_2 that accounts for about 2.5 Mt CO_2 in 2019 with an emission factor of 0.785 tonnes of CO_2 per tonne of calcium oxide (CaO), a primary constituent of cement, produced (IEA, 2021d, United States Congress House Committee on Science – Subcommittee on Energy and Environment, 2000). Post-combustion technologies can be applied to cement production plants to capture CO_2 produced (about 15% to 30% by volume (vol.) in the flue gases) from the calcination of limestone, (calcium carbonate, $CaCO_3$), which was driven by the combustion of fossil fuel (Metz et al., 2005). Oxy-fuel combustion capture system can also be employed in a cement plant by substituting air in the cement kiln with pure O_2, which would increase CO_2 concentration in the flue gases to 70% or higher (Beumelburg, 2021). These approaches are being developed at HeidelbergCement's Norcem Brevik plant in Norway, which is a 0.4 Mt/year CO_2 capture facility that is currently in construction (Turan et al., 2021). A breakthrough, low-cost carbon capture technology enabling the cement and lime industries to capture CO_2 from raw limestone based on Calix's new process of 'indirect calcination' is being developed by the LEILAC (Low Emissions Intensity Lime and Cement) project in Belgium. The LEILAC process involves heating the limestone through a special steel tube within the 'pre-calciner'

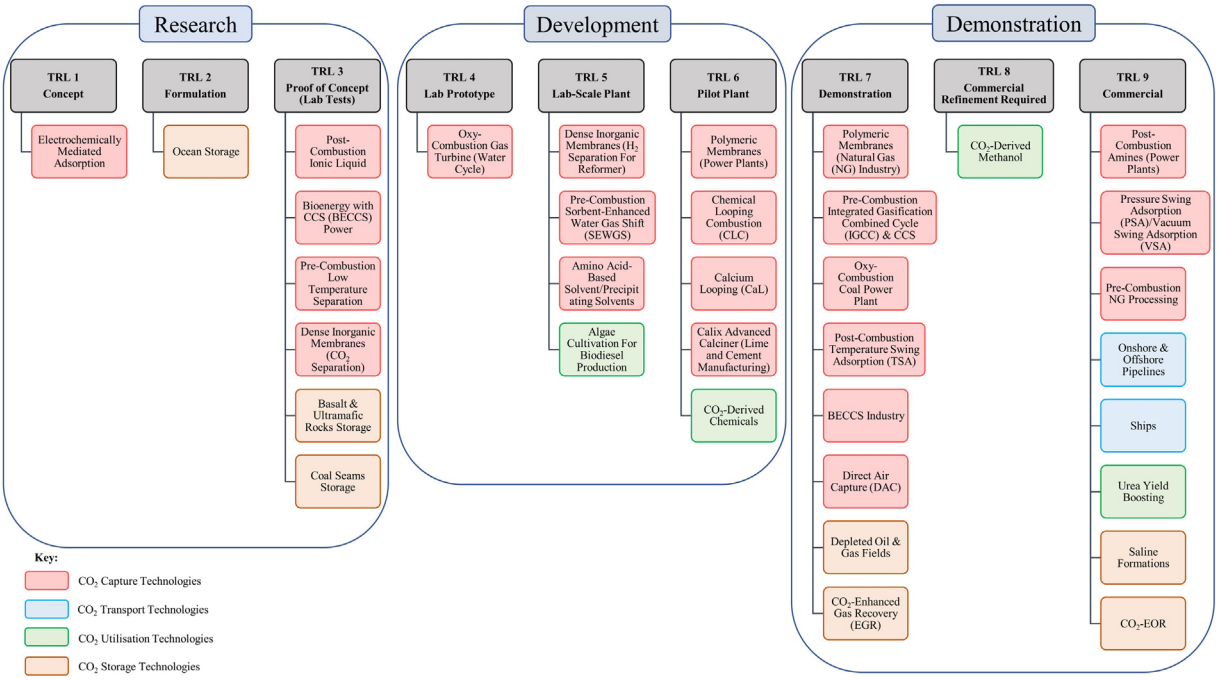

Figure 4. Technology readiness level (TRL) of some CCUS technologies. Adapted from (Bui et al., 2018; Kearns et al., 2021).

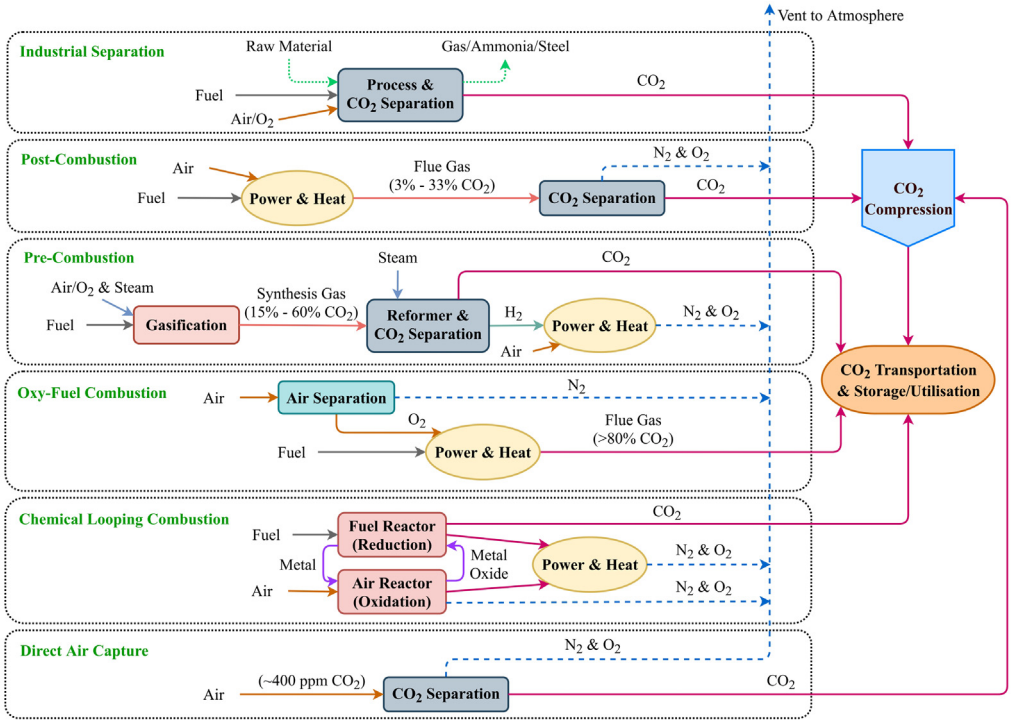

Figure 5. Schematic diagram of current CO_2 capture technologies (Metz et al., 2005; Wang and Song, 2020).

so that pure CO_2 can be captured as it is released from the limestone and kept separated from the furnace exhaust gases (Project LEILAC, 2021).

Aluminium production is a highly energy-intensive industry with large electricity needs (accounting for about 3.5% of global electricity consumption) (Gomilšek et al., 2020) and process emissions (accounting for about 2.5 Gt CO_2 globally in 2020) (IEA, 2020b). In aluminium smelting process (also known as the Hall-Héroult process), the carbon anodes are oxidised when alumina (Al_2O_3) is reduced to aluminium metal during electrolysis and the CO_2 formed is ducted away from the smelting pots with fresh air, producing a very low concentration of CO_2 in exhaust gases, which is a challenge for employing CCS technologies to aluminium smelters (Turan et al., 2021). There have been ongoing innovation efforts to develop inert anodes that are made from alternative materials (for example, a ceramic composite (Nature, 2018)), do not degrade and produce pure O_2 instead of CO_2 (IEA, 2021e).

3.1.2. Post-combustion capture

Post-combustion capture involves separation and capture of CO_2 from flue gas produced from the combustion of fossil fuels in air. This technology is mature and it can be applied to existing and future power

Table 2
Advantages and disadvantages of current CO_2 capture technologies (Wang and Song, 2020; Leung et al., 2014).

Capture Technologies	Advantages	Disadvantages
Industrial Separation	• Low-cost CO_2 capture for industrial processes with high CO_2 concentration • Commercially deployed in some industries	• Low CO_2 concentration in aluminium production makes the process energy-intensive
Post-Combustion	• More mature technology compared to other alternatives • Can easily retrofit to existing plants	• Low CO_2 concentration and partial pressure reduce capture efficiency • High energy requirement • High capital and operating costs
Pre-Combustion	• High CO_2 concentration and partial pressure enhance sorption efficiency • Fully developed technology, commercially deployed in some industries • Possible for retrofit to existing plants	• Temperature (associated with heat transfer) and efficiency issues (associated with H_2-rich gas turbine fuel) • High energy requirement for sorbent regeneration • High capital and operating costs for current sorption systems
Oxy-Fuel Combustion	• Very high CO_2 concentration enhances absorption efficiency • Mature air separation technologies available • Possibility of compact boiler and other equipment with reduced volume of flue gas to be treated	• High efficiency drop • Costly and energy-intensive O_2 production • May have corrosion problem
Chemical Looping Combustion (CLC)	• Energy-intensive air separation can be prevented as very high CO_2 concentration can be obtained from the process • Low-cost O_2 carrier materials	• Currently under development • Fuel must be desulphurised before entering the fuel reactor to prevent formation of metal sulfides.
Direct Air Capture (DAC)	• Directly reduce atmospheric CO_2 concentration • High CO_2 removal efficiency	• Extremely low CO_2 concentration (~400 ppm) in air makes the process costly and energy-intensive

Table 3
CO_2 removal efficiency, energy consumption and cost of CO_2 capture of different CO_2 capture technologies.

CO_2 Capture Technology	CO_2 Removal Efficiency (% vol.)	Energy Consumption for CO_2 Capture (GJ/t CO_2)	CO_2 Capture Cost (USD/t CO_2)	References
Industrial Separation	90	5.00	34.80 to 60.90	(Idem et al., 2015; Romeo et al., 2008)
Post-Combustion	90	4.14	46 to 74	(Nasr et al., 2015; Rubin et al., 2015)
Pre-Combustion	90	3.35	34 to 63	(Rubin et al., 2015; Dinca et al., 2018; Kato et al., 2005)
Oxy-Fuel Combustion	>90	4.05	52	(Rubin et al., 2015; Vega et al., 2019; Senior et al., 2013)
Chemical Looping Combustion (CLC)	96 to 99	0.95	<59.20	(Markström et al., 2013; Zhou et al., 2021; Oh et al., 2021)
Direct Air Capture (DAC)	85 to 93	5.25	140 to 340	(Deutz and Bardow, 2021; Keith et al., 2018; Abanades et al., 2020)

plants (Younas et al., 2016). Generally, flue gas stream has a low CO_2 concentration (3% to 33% vol.) and a low CO_2 partial pressure (0.03 bar to 5 bar) (Wang and Song, 2020) with a high flow rate (5 to 10 times larger than typical streams for CO_2 removal in natural gas and chemical industries) (Merkel et al., 2010). Besides inert gases (such as nitrogen (N_2), O_2 and H_2O vapour), flue gases also contain air pollutants such as sulphur dioxide (SO_2), nitrogen oxides (NO_x), particulates (fly ash), trace metals and other traces of inorganic and organic contaminants that need to be removed and/or reduced to a very low level prior to CO_2 capture process (Metz et al., 2005; Merkel et al., 2010). All these factors elevated the energy penalty and associated costs for the capture unit to reach CO_2 concentration above 95.5% vol. for transport and storage (Koukouzas et al., 2020; de Visser et al., 2008).

In addition, the performance and cost of power plants with CCS vary depending on the fuel sources and CO_2-generating processes. For post-combustion capture (CO_2 capture efficiency assumed to be 90%) at new natural gas combined cycle (NGCC) power plants, the overall plant energy consumption per MWh of net electricity would increase by an average of 16% (from 13% to 18%) and the cost of electricity per MWh would increase by an average of 50% (from 28% to 72%) while that at supercritical pulverized coal (SCPC) power plants using bituminous coals, the overall plant energy consumption per MWh of net electricity would increase by an average of 32% (from 21% to 44%) and the cost of electricity per MWh would increase by an average of 73% (from 48% to 98%) (Rubin et al., 2015). Kheirinik *et al.* (2021) reported that post-combustion at pulverized coal power plants are more cost-effective (levelized cost of electricity, LCOE, of GBP 142/MWh, i.e., about USD 192/MWh) than pre-combustion capture at IGCC power plants (LCOE of GBP 148.06/MWh, i.e., about USD 200/MWh) due to its low capital and indirect costs (Kheirinik et al., 2021). Several CO_2 separation

technologies such as absorption, adsorption, membrane, biological and cryogenic can be applied for post-combustion capture and they are further described in Section 3.2.

3.1.3. Pre-combustion capture

Pre-combustion capture involves capturing CO_2 prior to fuel combustion (Eide and Bailey, 2005). This technology can be applied to NGCC and integrated gasification combined cycle (IGCC) power plants and it is based on processes used industrially for the production of hydrogen and chemical commodities. Pre-combustion CO_2 capture technology in the chemical industry is mature and has been used for over 95 years. The fuel (coal or natural gas) is pre-treated by reacting it with oxygen or air and/or steam to produce synthesis gas (or syngas) (containing CO and H_2) via gasification for coal or reforming (either steam reforming (Equation 1) or partial oxidation (Equation 2)) for natural gas. The produced CO reacts with steam in a catalytic reactor (or shift converter) to form CO_2 and more H_2 via water-gas shift (WGS) reaction (Equation 3) (Jansen et al., 2015).

$$\text{Steam reforming}: \ CH_4 + H_2O \rightleftharpoons CO + 3H_2 \ \Delta H^0_{298} = 206 \text{ kJ/mol} \quad (1)$$

$$\text{Partial oxidation}: \ CH_4 + \frac{1}{2}O_2 \rightleftharpoons CO + 2H_2 \ \Delta H^0_{298} = -36 \text{ kJ/mol} \quad (2)$$

$$\text{WGS reaction}: \ CO + H_2O \rightleftharpoons CO_2 + H_2 \ \Delta H^0_{298} = -41 \text{ kJ/mol} \quad (3)$$

The CO_2 (typically concentration of 15% to 60% vol. on a dry basis and total pressure of 2 MPa to 7 MPa) can then be separated from H_2, commonly by physical/chemical absorption process, resulting in an H_2-rich fuel gas that can be utilised for power and heat generation such as boilers, gas turbines, engines and fuel cells (Metz et al., 2005). The high CO_2 concentration and partial pressure in the flue gas enhance the sorption efficiency, making the CO_2 separation and compression process less energy demanding than post-combustion capture processes (Leung et al., 2014). Gazzani *et al.* (2013) reported that an improvement of 60% in sorbent capacity for CO_2 capture in NGCC power plants with sorption enhanced WGS could reduce the energy consumption for CO_2 avoided from 2.9 MJ/kg CO_2 to 2.5 MJ/kg CO_2, which was 26% and 19% lower than the NGCC power plants with CO_2 capture via amine scrubbing using monoethanolamine (MEA) and methyldiethanolamine (MDEA) solvents, respectively (Gazzani et al., 2013). Moreover, energy is also required for air separation, gasification/reforming and improvements in energy recovery during syngas temperature swing (Jansen et al., 2015).

For pre-combustion capture at new IGCC power plants with bituminous coal, the energy requirement for CO_2 capture was lower by 10%/MWh but their CO_2 capture cost was higher by USD 83/t CO_2 compared to SCPC power plants without CCS (Rubin et al., 2015). Many studies tend to focus on pre-combustion CO_2 capture on coal-based power generation plants than on natural gas-based power generation plants because the plant complexity and costs in the latter case are not competitive with post-combustion CO_2 capture on natural gas-based power generation plants (Kanniche et al., 2010). Pre-combustion capture can be achieved by several CO_2 separation technologies such as absorption, adsorption, membrane and cryogenic, which will be described more in Section 3.2.

3.1.4. Oxy-fuel combustion capture

Oxy-fuel combustion capture involves combustion of fuel in a nearly pure O_2 (95% to 99% vol.) at very high temperature (typically about 1,300 °C to 1,400 °C in a gas turbine cycle and about 1,900 °C in an oxy-fuel coal-fired boiler) to produce flue gas consisting of CO_2 (>80% vol.), H_2O vapour, particulate matter (fly ash) and traces of contaminants (such as SO_2 and NO_x) (Metz et al., 2005, Rubin et al., 2012). Particulates can be removed by conventional electrostatic precipitator while SO_2 and NO_x can be removed by flue gas desulphurisation and denitrification methods (Leung et al., 2014). The H_2O vapour can be removed by cooling and compressing the gas stream (Rubin et al., 2012). CO_2 can easily be separated from the flue gas without the need for chemical solvent or physical sorbent due to the high concentration in the flue gas stream, which can then be compressed, transported and stored. Among available CO_2 capture technologies, oxy-fuel combustion capture is the most promising energy-efficient technology for CO_2 capture with a low-efficiency penalty of 4% compared with 8% to 12% for the post-combustion capture technology even though it is yet to be industrially applicable (Wienchol et al., 2020).

The main challenge in oxy-fuel combustion capture is the cryogenic air separation process that requires an air separation unit to generate nearly pure O_2 for combustion, which is energy-intensive and costly. The air separation unit (ASU) was estimated to consume up to 15% of the power plant's electrical output and represent about 26% of the total equipment costs (Herzog and Golomb, 2004; López et al., 2016). New air separation methods such as oxygen- and ion-transport membranes or chemical looping have been investigated and they showed capability in producing inexpensive O_2 for oxy-fuel combustion process (Falkenstein-Smith et al., 2017). It has been found that oxy-fuel combustion process with an oxygen-transport membrane (OTM) unit could save the energy efficiency by 0.5% to 9% for pulverized coal combustion with air and by 0.3% to 2.9% for IGCC power plant compared to oxy-fuel combustion process with ASU (Portillo et al., 2019). An economic saving of 33% in overall plant costs could be achieved from coal-fired boilers with oxy-fuel combustion and OTM unit compared to a coal-fired boiler with post-combustion and MEA scrubbing (Carbo et al., 2009). On the other hand, chemical looping air separation (CLAS) based on chemical looping with oxygen uncoupling (CLOU) offers about 40% to 70% lower operational costs and substantially lower capital investment than the conventional cryogenic air separation method (Zhou et al., 2016).

Oxy-fuel combustion capture can be employed in existing or new power plants using various types of fuels such as municipal solid waste or lignocellulosic biomass to produce bioenergy and biogenic CO_2. This bioenergy with CCS (BECCS or BioCCS) process is one of the best options to achieve net-zero or negative CO_2 emissions (Wienchol et al., 2020). Rosa *et al.* (2021) estimated about 200 Mt biogenic CO_2/year could be removed in Europe from existing point sources (31% from pulp and paper facilities, 18% from waste-to-energy facilities, 15% from biomass co-fired plants and 1% from wastewater treatment plants) and distributed sources (17% from crop residues, 9% from livestock manure and 9% from household organic food waste) (Rosa et al., 2021). Most of the BECCS facilities in the world involve the capture of CO_2 from ethanol production plants for EOR (Shahbaz et al., 2021). The cost of implementing BECCS technology varies widely depending on the type of industry (between USD 88/t CO_2 avoided and USD 288/t CO_2 avoided from biomass combustion, between USD 20/t CO_2 avoided and USD 175/t CO_2 avoided from ethanol, between USD 20/t CO_2 avoided and USD 70/t CO_2 avoided from pulp and paper mills and between USD 30/t CO_2 and USD 76/t CO_2 avoided from biomass gasification) (Consoli, 2019). Wei *et al.* (2020) reported that BECCS technology will be more economically feasible than fossil fuel with CCS if the carbon tax is higher than USD 28.30/t CO_2 (Wei et al., 2020).

3.1.5. Chemical looping combustion (CLC) capture

CLC is an emerging technology for power plants and industrial applications with inherent CO_2 capture of minimal energy penalty and cost for the separation process. CLC involves oxidising the fuel with a solid O_2 carrier (typically a metal oxide (MeO)) instead of air. The CLC process consists of two interconnected reactors: a fuel reactor (FR), where fuel is oxidised by the O_2 carrier to produce CO_2 and H_2O vapour (Equation 4), and an air reactor (AR), where the reduced O_2 carrier (i.e., a metal (Me)) is oxidised by the air (Equation 5). The H_2O vapour is condensed to obtain nearly pure CO_2 from the FR flue gas that is ready for storage (Moldenhauer et al., 2020).

$$\text{Reduction}: \ 4MeO + CH_4 \rightarrow 4Me + 2H_2O + CO_2 \quad (4)$$

$$\text{Oxidation}: \ 4Me + 2O_2 \rightarrow 4MeO \quad (5)$$

Although CLC is an attractive technology, it is still under development as this process is relatively more complicated than the oxy-combustion process (IEAGHG, 2014). The CLC process was first developed for gaseous fuels (such as natural gas and syngas), which then extended to solid fuels (such as coal and biomass) and later to liquid fuels (such as oil and ethanol) (Abuelgasim et al., 2021). A recent study showed that NGCC with CLC process with an air inlet to a gas turbine and steam turbine integration is the most efficient process for generating power from natural gas as it has a higher efficiency (44.3%) and a lower CO_2 capture cost (USD 59.20/t CO_2) than NGCC with post-combustion capture using MEA (power plant efficiency of 43.8% and CO_2 capture cost of USD 76.20/t CO_2) (Oh et al., 2021). Zhao *et al.* (2021) reported that coal-based CLC power plant was more sustainable and has a lower total life cycle cost (LCOE of USD 0.14/kWh) than other conventional coal-based power plants with CCS (LCOE of USD 0.19/kWh for IGCC power plant with CCS using Selexol, USD 0.18/kWh for oxy-fuel combustion power plant using distillation and LCOE of USD 0.17/kWh for ultra-supercritical (USC) power plant with CCS using MEA) (Zhao et al., 2021). Research on CLC with liquid fuels is limited, mainly due to inexperienced utilisation of liquid with a fluidized bed reactor (Abuelgasim et al., 2021). One of the methods is by injecting liquid fuels directly to the fluidized bed FR. Diego *et al.* (2016) demonstrated that ethanol has the capacity to be used as fuel in a 1 kilowatt-thermal (kW_{th}) continuous CLC unit, achieving almost 100% CO_2 capture efficiency with O_2 carriers based on 14% by weight (% wt.) copper impregnated on gamma-alumina (γ–Al_2O_3) (Cu14–γAl) and on 18% wt. nickel impregnated on alpha-alumina (α–Al_2O_3) (Ni18–αAl) at normal operating conditions (de Diego et al., 2016).

3.1.6. Direct air capture (DAC)

Besides BECCS, DAC is another negative emissions technology. DAC involves extracting CO_2 directly from the atmosphere and this innovative technology is in the early demonstration stage (TRL 7). Currently, there are two commercial pathways for DAC, which are liquid solvent-based and solid sorbent-based approaches. In liquid solvent-based approach, a very dilute CO_2 (~400 ppm in the atmosphere) is first absorbed into a strong alkaline liquid solvent such as aqueous calcium hydroxide ($Ca(OH)_2$) solution in an adsorption column/contactor to form $CaCO_3$ (Equation 6), which can then be separated, dried and calcined at 900 °C to form CaO and release concentrated CO_2 (Equation 7). CaO is hydrated in a slaker unit for $Ca(OH)_2$ regeneration (Equation 8) (Sanz-Pérez et al., 2016).

$$\text{Absorption}: \ Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \ \Delta H^0_{298} = -109 \text{ kJ/mol} \quad (6)$$

$$\text{Calcination}: \ CaCO_3 \rightarrow CaO + CO_2 \ \Delta H^0_{298} = 179.2 \text{ kJ/mol} \quad (7)$$

$$\text{Slaking}: \ CaO + H_2O \rightarrow Ca(OH)_2 \ \Delta H^0_{298} = -64.5 \text{ kJ/mol} \quad (8)$$

Although this approach allows adequate CO_2 separation, the process has a high energy penalty for $Ca(OH)_2$ regeneration of about 15 GJ/t CO_2 and this will make the process costly to operate (Baciocchi et al., 2006). Other liquid solvents with strong binding of CO_2 such as aqueous solutions of sodium hydroxide (NaOH) and potassium hydroxide (KOH) have also been used as alternatives (Fasihi et al., 2019). Depending on the design choices of the gas-liquid contactor, the cost of the air contactor can range from USD 60/t CO_2 for a contactor design based on a closed counter-flow gas scrubber column to USD 240/t CO_2 for a contactor design based on a cooling tower that consisted of an open contactor with cross-flow and a slab geometry (Holmes and Keith, 2012).

A recent study reported that the cost of a DAC system based on passive CO_2 carbonation of porous $Ca(OH)_2$ plates was estimated to be between USD 140/t CO_2 and USD 340/t CO_2 (Abanades et al., 2020). The DAC cost can be reduced using low-cost polyvinyl chloride (PVC) gas-liquid contactors. For example, Keith *et al.* (2018) estimated a lower CO_2 capture cost between USD 94/t CO_2 and USD 232/t CO_2 for a 1 Mt CO_2/year DAC plant based on aqueous NaOH sorbent coupled to a calcium caustic recovery loop with structured PVC packing (Keith et al., 2018). These cost estimates for DAC exceed that of all the other CO_2 capture pathways mentioned earlier in this section (refer to Table 3) and this was primarily attributed to the very low concentration of CO_2 to be captured from the air, which is about 300 times more dilute than from a typical flue gas stream (Azarabadi and Lackner, 2019).

In solid sorbent-based approach, CO_2 is adsorbed onto a solid sorbent such as zeolites, activated carbons, metal-organic frameworks (MOFs), amine-modified materials, silica materials, porous organic polymers, carbon nanotubes and carbon molecular sieves (McQueen et al., 2021). This approach has a lower energy consumption (as low as 1 GJ/t CO_2) due to the low temperatures (between 80 °C and 100 °C) for solid sorbents regeneration, which would lead to reduced CO_2 capture cost of the DAC plant (Fasihi et al., 2019; Sinha and Realff, 2019). Sinha et al. (2019) estimated that the cost of DAC using solid adsorbents was between USD 86/t CO_2 and USD 221/t CO_2, which was lower than the cost estimates mentioned earlier for DAC systems using liquid solvents (Sinha and Realff, 2019). Other pathways for DAC have also been proposed but they are not yet widely studied, for examples, electrochemical approaches, mineral carbonation, membranes and photocatalytic CO_2 conversion (Sanz-Pérez et al., 2016).

3.2. CO_2 separation

There are several CO_2 separation technologies, which include: (i) absorption, (ii) adsorption, (iii) membrane, (iv) cryogenic and (v) biological (Figure 6). The advantages and disadvantages of these CO_2 separation technologies are summarised in Table 4. Additionally, their key technical and economic parameters (such as CO_2 removal efficiency, energy consumption for CO_2 capture and cost of CO_2 capture) are provided in Table 5, which are useful information when making the choice of CO_2 separation technology for a particular CCS project besides the operational parameters and gas composition of the gas stream to be treated. These CO_2 separation technologies are described in the following sections (Sections 3.2.1 to 3.2.5).

3.2.1. Absorption

Absorption is the most mature and commercially available technology for separating CO_2 from the flue gas, particularly in petroleum and chemical industries, using a liquid absorbent (or solvent) that can be regenerated by heating (i.e., temperature swing) and/or depressurization (i.e., pressure swing) (Wang and Song, 2020; Leung et al., 2014). This technology can be divided into two classifications based on the nature of the interaction between a solvent and CO_2, namely: (i) chemical absorption that depends on the acid-base neutralisation reactions between an alkaline solvent and CO_2, and (ii) physical absorption that depends on the CO_2 solubility in an organic solvent and differences in temperature/pressure (Songolzadeh et al., 2014; Yuan and Eden, 2015). Chemical absorption can be used in post-combustion and pre-combustion capture systems while physical absorption is mainly used in the pre-combustion capture system.

The CO_2 absorption capacity, energy consumption and cost of CO_2 capture based on different chemical and physical solvents are compared in Table 6. The most common chemical solvents are aqueous alkanolamines that include primary amines (such as MEA and 2-amino-2-methyl-1-propanol (AMP)), secondary amines (such as diethanolamine (DEA)) and ternary amines (such as MDEA). On the other hand, the most common physical solvents are dimethyl ether of polyethylene glycol (DEPG) in the Selexol process, chilled methanol in the Rectisol process, N-methyl-pyrrolidone (NMP) in the Purisol process and propylene carbonate (PC) in the Fluor process. So far, the most efficient solvent for CO_2 absorption is MEA as it has high CO_2 absorption capacity (4.09 mol CO_2/kg solvent), high CO_2 recovery (typically between 85% and 90% vol.) and CO_2 purity above 99% vol. (Li et al., 2016; Yun et al., 2020; Xue et al., 2017). However, MEA-based CO_2 absorption has some

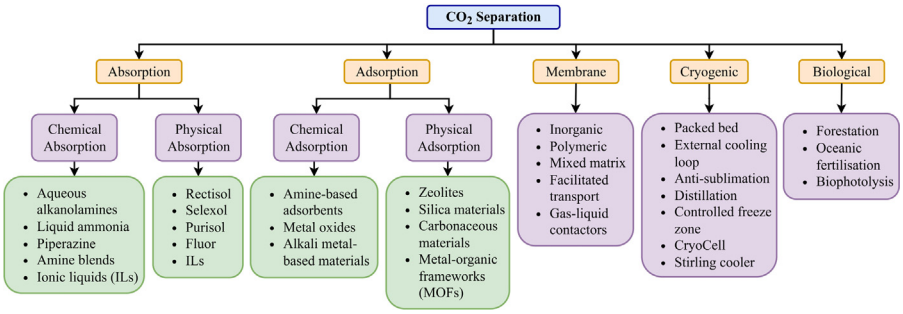

Figure 6. CO_2 separation technologies with some commonly used and emerging materials (Wang and Song, 2020; Ben-Mansour et al., 2016).

Table 4
Advantages and disadvantages of different CO_2 separation technologies (Metz et al., 2005; Wang and Song, 2020; Leung et al., 2014).

Separation Technologies	Advantages	Disadvantages
Absorption	• High absorption efficiency (>90% vol. CO_2) for concentrated CO_2 gas stream • Solvents can be regenerated by heating and/or depressurisation • High capacity at low temperature and high pressure • Low-cost solvent • Most mature technology	• Energy-intensive solvent regeneration • Solvents are corrosive and they can degrade by contaminants (e.g., sulphur oxides (SO_x) and NO_x) • High operating cost • Low capacity at high temperature and low pressure
Adsorption	• High adsorption efficiency (>85% vol. CO_2) • Reversible physical adsorption process • Adsorbent can be recycled, hence low waste generation • High capacity at low temperature and high pressure for physical adsorbents • High capacity at low CO_2 pressure for solid amine sorbents • Less corrosion	• Process using physical adsorbent has low CO_2 selectivity and capacity decreases with temperature and presence of moisture • Process using chemical adsorbents has high energy consumption due to high temperature requirement for CO_2 sorption and adsorbent regeneration • Solid amine sorbents degrade by thermal, oxidation and contaminants (e.g., SO_x and NO_x)
Membrane	• High separation efficiency (>80% vol. CO_2) • Relatively low operation cost • Easy handling and operation	• High manufacturing cost • Low permeability and fouling • Relatively low separation selectivity • May not stable in the presence of moisture
Cryogenic	• High separation efficiency (>85% vol. CO_2) • Mature technology	• Very energy-intensive process due to very low temperature and high pressure operation • Requires moisture pre-removal • May accumulate solidified CO_2 on the surface of heat exchanger
Biological	• No chemical hazards • High growth rate and CO_2 removal/fixation ability (up to 95% vol. by microalgae in closed ponds) • Co-production of food, biofuels and value-added products	• Time-consuming process • Requires large area • May affect biological diversity • Low separation efficiency (up to 40% vol. CO_2 using microalgae cultivated in open ponds) • Sensitive to contaminants (e.g., SO_x and NO_x) and culture conditions (pH, temperature and salinity)

drawbacks such as high energy consumption during solvent regeneration, solvent degradation (due to the presence of SO_2 and NO_2 in the flue gas) that could lead to solvent loss and evaporation, and high equipment corrosion rate (due to the presence of O_2), which makes the process costly to operate (Wang et al., 2011). For example, an ethylene glycol production process (from coal) with CO_2 capture based on MEA was estimated to have an energy consumption of 5.20 GJ/t CO_2 and a CO_2 capture cost of USD 39.38/t CO_2, the highest compared to the Rectisol process and that based on chilled ammonia and dimethyl carbonate (DMC) (Osagie et al., 2018).

Some possible alternatives to MEA solvent could be MDEA (as it has high CO_2 absorption capacity and low energy requirement), AMP (as it is less corrosive, has low degradation rate and low energy requirement), liquid ammonia (as it is less corrosive, high tolerance to particulate, SO_2, NO_x and O_2, and it has low energy requirement), piperazine (PZ) (as it is less corrosive, has low energy requirement, fast CO_2 absorption rate and low degradation rate) and blend of MDEA and PZ (MDEA/PZ) (as it has low energy requirement) (Ma et al., 2021; Zhuang et al., 2011; Zhao et al., 2017; Aroonwilas and Veawab, 2007; Freeman et al., 2009). For example, Otitoju *et al.* (2021) showed that 40% wt. PZ solvent-based post-combustion CO_2 capture process for flue gas from a 250-Megawatt

Table 5
CO_2 removal efficiency, energy consumption and cost of CO_2 capture of different separation technologies.

CO_2 Separation Technology	CO_2 Removal Efficiency (% vol.)	Energy Consumption for CO_2 Capture (GJ/t CO_2)	CO_2 Capture Cost (USD/t CO_2)	References
• Absorption (MEA)	85 to 90	3.80	62.80	(Li et al., 2016; Yun et al., 2020; Luis, 2016)
• Adsorption (13X zeolite)	88 to 95	1.17	89.66[a]	(Cheng et al., 2021; Zanco et al., 2021)
• Membrane (polymeric material)	70 to 90	1.28	80.46[a]	(Zanco et al., 2021; Zhai and Rubin, 2013)
• Cryogenic (packed bed)	90 to 99.99	1.80	52[d]	(Leung et al., 2014; Song et al., 2019; Tuinier et al., 2011; Song et al., 2012)
• Biological (microalgae)	10 to 40[b]; up to 95[c]	3.09[b]; 8.31 – 70.36[c]	793.00[c]	(Rezvani et al., 2016; Li et al., 2013; Jacob et al., 2015; Farrelly et al., 2013)

Notes:
[a] USD 1 = EUR 0.87.
[b] Microalgae cultivated in open ponds.
[c] Microalgae cultivated in closed ponds.
[d] Cost of cryogenic CO_2 capture using Stirling cooler system.

Table 6
CO_2 absorption capacity, energy consumption and cost of CO_2 capture based on different chemical and physical solvents.

Solvents	CO_2 Absorption Capacity (mol CO_2/ kg solvent)	Operating Temperature (°C)	Operating Pressure (bar)	Energy Consumption for CO_2 Capture (GJ/t CO_2)	CO_2 Capture Cost (USD/t CO_2)	References
Chemical solvents						
2-amino-2-methyl-1-propanol (AMP) (30% wt.)	2.24	40	1	2.90	69.30	(Ma et al., 2021)
Diethanolamine (DEA) (40% wt.)	0.95	40	1	1.59	26.8[a]	(Xue et al., 2017; Wang et al., 2011; Osagie et al., 2018; Ma et al., 2021; Yakub et al., 2014)
Liquid ammonia (11.7%)	1.91	10	1	3.64	36.57[b]	(Osagie et al., 2018; Zhuang et al., 2011)
Methyldiethanolamine (MDEA) (20% wt.)	8.38	20	1	3.26	52.10[c]	(Santos et al., 2016; Cormos, 2015)
Monoethanolamine (MEA) (30% wt.)	4.09	40	1	5.20	39.38[b]	(Xue et al., 2017; Osagie et al., 2018)
Piperazine (PZ) (40% wt.)	2.38	40	0.05	2.76	34.65	(Rochelle et al., 2011; Otitoju et al., 2021)
MDEA/PZ blend (30:20, % wt.)	0.49	40	1.1	2.24	50.88[d]	(Zhao et al., 2017; Chauvy et al., 2021)
Task-specific ionic liquids (ILs)	2.66[e]	20	1	1.40[f]	62.40[g]	(Luo et al., 2014; de Riva et al., 2018; Zhai and Rubin, 2014)
Physical solvents						
Methanol (Rectisol)	0.21	–30	24	1.88	26.65[b]	(Osagie et al., 2018; Descamps et al., 2008)
Dimethyl carbonate (DMC)	0.28	25	1	2.10	30.37[b]	(Osagie et al., 2018; Zhao et al., 2018)
Dimethyl ether of polyethylene glycol (DEPG) (Selexol)	0.14	25	1	7.54	7.46[h]	(Ramdin et al., 2012; Li et al., 2020; Ashkanani et al., 2020)
Conventional ILs	2.23[i]	40	27.4	1.40[j]	16.53[k]	(de Riva et al., 2018; Ashkanani et al., 2020; Zhang et al., 2012)
N-methyl-pyrrolidone (NMP) (Purisol)	0.14	25	1	9.93	18.27[l]	(Ramdin et al., 2012; Li et al., 2020; Ashkanani et al., 2020)
Propylene carbonate (PC) (Fluor)	0.12	25	1	8.30	54.13	(Ramdin et al., 2012; Li et al., 2020)

Notes:
[a] USD 1 = GBP 0.74.
[b] USD 1 = CNY 6.36.
[c] USD 1 = EUR 0.87.
[d] CO_2 absorption in 10% wt. aqueous MDEA and 30% wt. PZ blend solution at 40 °C and 1.2 bar.
[e] CO_2 absorption in 4-azabenzimidazole-based IL ($[P_{66614}][4\text{-ABI}]$).
[f] Energy consumption for CO_2 capture with the triethyl(octyl)phosphonium 2-cyanopyrrolide ($[P_{2228}][CNPyr]$) and the trihexyltetradecylphosphonium 2-cyanopyrrolide ($[P_{66614}][2\text{-CNPyr}]$) Aprotic Heterocyclic Anion (AHA) ILs.
[g] CO_2 capture cost with ($[P_{66614}][2\text{-CNPyr}]$-based IL.
[h] CO_2 capture cost with solvent at 5 °C.
[i] CO_2 absorption in trihexyl(tetradecyl)phosphonium bis(trifluoromethylsulfonyl)imide-based IL ($[P_{14,6,6,6}][Tf_2N]$).
[j] Energy consumption for CO_2 capture with imidazolium-based ILs.
[k] CO_2 capture cost using 1-Hexyl-3-methyl-imidazolium bis(trifluoromethylsulfonyl)imide-based IL ($[hmim][Tf_2N]$) at 0 °C.
[l] CO_2 capture cost with solvent at 0 °C.

(MW) NGCC power plant would bring technical and economic benefits (low energy demand of 2.76 GJ/t CO_2 with an advanced flash stripper and reduced CO_2 capture cost of USD 34.65/t CO_2) compared to the current 30% wt. MEA solvent-based process, which has a higher energy demand of 5.34 GJ/t CO_2 and cost of CO_2 capture at USD 61.13/t CO_2 (Otitoju et al., 2021).

Recently, ionic liquids (ILs) are considered as potential alternatives to traditional absorbents due to their low volatility, good thermal stability and very low energy requirement for regeneration (Wang and Song, 2020). The CO_2 absorption capacity of ILs varied widely, from an average of 6.03 mol CO_2/kg IL (between 0.06 mol CO_2/kg IL and 12 mol CO_2/kg IL) for chemical absorption of CO_2 by conventional (amine-free functionalized) ILs or ionic salts with single active site to an average of 8.84 mol CO_2/kg IL (between 0.66 mol CO_2/kg IL and 8.18 mol CO_2/kg IL) for chemical absorption of CO_2 by task-specific (amino-functionalized) ILs or ionic salts with a single active site and an average of 8.65 mol CO_2/kg IL (between 1.53 mol CO_2/kg IL and 7.12 mol CO_2/kg IL) for chemical absorption of CO_2 by multiple active sites functionalized ILs and functionalized IL-based blends (Cui et al., 2016). For ILs with physical absorption, the CO_2 absorption capacity was about 2.10 mol CO_2/kg IL on average (between 0.45 mol CO_2/kg IL and 3.75 mol CO_2/kg IL) (Zhang et al., 2012). The main challenge of ILs is that they are more expensive (higher by about 10 to 20) than conventional solvents and their price to performance ratio is not comparable with existing commercial solvents such as DEPG in Selexol process (Ramdin et al., 2012). More studies are still needed to develop solvents that are highly efficient, energy-saving and economical for large-scale CCS application.

3.2.2. Adsorption

Adsorption is another widely used industrial CO_2 separation technology that bound CO_2 gas molecules onto the surfaces of a solid adsorbent (Leung et al., 2014). This technology can be retrofitted to any power plant and applied to pre-combustion and post-combustion CO_2 capture systems as well as DAC system (Bui et al., 2018). Adsorption can operate at various temperature and pressure conditions, has high CO_2 removal efficiency (above 85% vol.) and high CO_2 purity (above 96% vol.), low energy requirement and low waste generation as adsorbents can be recycled (Leung et al., 2014; Cheng et al., 2021). This technology can be classified into two types, namely: (i) chemical adsorption that is driven by a chemical reaction between the solid adsorbent surface and CO_2, and (ii) physical adsorption that is driven by the interaction between the electric field of the solid adsorbent and the large quadrupole moment of CO_2 via physical forces such as van der Waals forces, electrostatic forces, dipole-dipole, apolar and hydrophobic interactions. Chemical adsorption can permanently bind and trap harmful gas on the solid surface and it is suitable for low CO_2 concentration gas streams while physical adsorption can temporarily bind gas molecules on the solid surface and it is normally applied to high CO_2 concentration gas streams (Younas et al., 2016; Ho et al., 2014).

A variety of solid adsorbents have been studied for CO_2 capture. Amine-based sorbents, metal oxides and alkali metal-based materials are some common types of chemical adsorbents whereas zeolites, silica materials, carbonaceous materials and MOFs are some common types of physical adsorbents. The CO_2 adsorption capacity and operating conditions for selected chemical and physical absorbents are provided in Table 7. Solid amine-based sorbents have been widely studied due to their low heat of regeneration (resulting from the low heat capacity of the solid supports) compared with aqueous amines. However, their commercial usage is challenging because of their low CO_2 adsorption capacity and high production cost (Yu et al., 2012). Several approaches have been attempted to improve the CO_2 capture properties of amine-based sorbents, which include preparing support materials with high amine contents, using amines with high nitrogen content and developing effective methods for amine immobilization (Wang et al., 2011). For example, the highest CO_2 adsorption capacity of amine-based sorbents found in the literature was 21.45 mol CO_2/kg adsorbent using 70% wt. tetraethylenepentamine (TEPA) impregnated on poly(methyl methacrylate) (PMMA) support (TEPA-PMMA) at 23 °C and 1 bar with feed gas composition containing 15% vol. CO_2 and 2.6% vol. H_2O vapour in N_2 (Lee et al., 2008). Generally, amine-based sorbents can offer high CO_2 capacity and selectivity, fast kinetics, tolerance to H_2O, stability for multicycle, no or less corrosion on equipment and low regeneration energy (Wang and Song, 2020).

Metal oxides and metal-oxide based adsorbents are popular as pre-combustion CO_2 adsorbents because of their low cost, abundance, low toxicity, durability over repeated cycles and able to operate at medium to high temperatures (200 °C to 500 °C for magnesium oxide (MgO) and 500 °C to 900 °C for CaO) despite having slow reaction and requires high energy consumption for regeneration (Lee and Park, 2015; Huang et al., 2010; Bhatta et al., 2020). Alkali-metal based materials such as sodium carbonate (Na_2CO_3) are regenerable, low cost, has good sorption capacity and suitable for CO_2 capture from flue gases at low temperatures below 200 °C. However, they have some limitations for practical CO_2 capture application, which include carbonation reaction rate and temperature control, durability, slow heat mass transfer and irreversible reaction with contaminants in flue gas such as SO_2 and hydrogen chloride (HCl) (Wang et al., 2011; Kondakindi et al., 2013). For example, the heat transfer issue can be overcome by depositing carbonate-based sorbents onto a metal support such as Al_2O_3 that can quickly dissipate the heat to allow continuous adsorption-desorption cycles. Kondakindi *et al.* (2013) reported a high adsorption capacity of 7.70 mol CO_2/kg adsorbent for 35% wt. Na_2CO_3/Al_2O_3 at 45 °C and the samples were considered durable with a capacity loss between 40% to 50% with 500 cycles (Kondakindi et al., 2013).

Another type of chemical adsorbents is alkali metal-based materials such as lithium zirconate (Li_2ZrO_3) and lithium silicate (Li_4SiO_4). Although this material has high CO_2 adsorption capacity (≤6.5 mol CO_2/kg adsorbent), small volume change during adsorption-desorption cycles and they can be used for direct CO_2 separation from flue gas at high temperatures (400 °C to 600 °C), they have kinetic limitation (Wang et al., 2011; Ochoa-Fernández et al., 2005). This material is not well investigated and more research to improve its kinetic properties for CO_2 capture is required.

Besides chemical adsorbents, physical adsorbents such as zeolites, silica materials, carbonaceous materials and MOFs can also be used for CO_2 separation at low temperature (<200 °C) (Wang et al., 2011). Zeolites are widely used in the refinery and gas separation industry due to their high CO_2 adsorption capacity, especially 13X zeolite that could adsorb as much as 6.18 mol CO_2/kg adsorbent at 25 °C and 1 bar (Wang and Song, 2020; Chen et al., 2014). However, they have several drawbacks, which include low selectivity of CO_2 over other gases (such as N_2, methane (CH_4), H_2O and etc.), rapid decline in adsorption capacity with increasing temperature above 30 °C and become negligible above 200 °C as well as in the presence of moisture in the gas stream, and requirement for high regeneration temperature (often >300 °C) (Yu et al., 2012; Xu et al., 2003). Silica materials are mostly studied as support materials for amine-based sorbents or metal-based materials because of their high surface area and pore volume, tunable pore size and good thermal and mechanical stability despite low CO_2 adsorption capacity (Yu et al., 2012). Both zeolites and silica materials have low production costs (Lee and Park, 2015).

Carbonaceous materials such as activated carbon have been widely used for CO_2 capture due to their high thermal stability, tolerance to H_2O vapour, abundance and low cost. Their adsorption/desorption temperatures are below 100 °C and they can be used at atmospheric pressure. The drawbacks of carbonaceous materials are their high sensitivity to temperature and relatively low selectivity for CO_2 over other gases (Yu et al., 2012; Lee and Park, 2015). One approach to improve the CO_2 adsorption capacity and selectivity is to increase the surface area and tune the pore structure of the carbonaceous adsorbents using different precursors or fabricating different structures such as single-

Table 7
CO_2 adsorption capacity and operating conditions for selected chemical and physical adsorbents.

Adsorbent Type	Adsorbents	CO_2 Adsorption Capacity (mol CO_2/kg adsorbent)[a]	Operating Temperature (°C)	Operating Pressure (bar)	References
Chemical adsorbents					
Amine-based sorbents	Diethylenetriaminopropyl (DT(NNN)) (10 SiOH/nm^2 SBA-15)-SBA-15	1.82	45	1	(Sanz et al., 2012)
	Diethylenetriamine (DETA) (40% wt.)-MCM-41	1.43[b]	35	1	(Liu et al., 2015)
	Ethylenediamine (EDA) (40% wt.)-MCM-41	1.19[b]	35	1	(Liu et al., 2015)
	Pentaethylenehexamine (PEHA) (40% wt.)-MCM-41	2.34[b]	35	1	(Liu et al., 2015)
	Branched-polyethylenimine (BPEI) (Molecular weight = 800 Da) (BPEI/800) (60% wt.)-nano-silica	4.59	105	1	(Li et al., 2014)
	Polyethylenimine (PEI) (60% wt.)- hexagonal mesoporous silica (HMS)	4.18	75	–	(Chen et al., 2010)
	PEI (50% wt.)-SBA-15	1.69	45	1	(Sanz et al., 2012)
	Tetraethylenepentamine (TEPA) (40% wt.)-MCM-41	1.96[b]	35	1	(Liu et al., 2015)
	TEPA (70% wt.)-poly(methyl methacrylate) (PMMA)	21.45[c]	23	1	(Lee et al., 2008)
	Triamine-containing silane (TRI)-pore-expanded (PE)-MCM-41	2.85	25	1	(Serna-Guerrero et al., 2010)
Metal oxides	Calcium oxide (CaO)	9.31[d]	25	1	(Graados-Pichardo et al., 2020)
	Mesoporous iron oxide (Fe_2O_3)	0.15	0	1	(El-Desouky et al., 2020)
	Mesoporous magnesium oxide (MgO)	1.82[e]	25	1	(Bhagiyalakshmi et al., 2020)
Alkali metal-based materials	Lithium orthosilicate (Li_4SiO_4)	5.66	746	1	(Domenico et al., 2019)
	Lithium zirconate (Li_2ZrO_3)	5.00	600	1	(Ochoa-Fernández et al., 2005)
	Sodium carbonate (Na_2CO_3) (35% wt.)-aluminium oxide (Al_2O_3)	7.70[f]	45	–	(Kondakindi et al., 2013)
Physical adsorbents					
Zeolites	13X zeolite	6.18	25	1	(Chen et al., 2014)
	5A zeolite	3.38	30	1	(Liu et al., 2011)
	Clinoptilolite (ZN-19)	0.04	17	0.26	(Hernández-Huesca et al., 1999)
	Erionite (ZAPS)	0.07	17	0.26	(Hernández-Huesca et al., 1999)
	Fly ash-derived zeolite[g]	4.16	50	1	(Czuma et al., 2020)
	H-ZSM-5-30	0.04	22	1	(Harlick and Tezel, 2004)
	Mordenite (ZNT)	0.04	17	0.26	(Hernández-Huesca et al., 1999)
Silica materials	MCM-41	0.62	25	1	(Serna-Guerrero et al., 2010)
	PE-MCM-41	0.50	25	1	(Serna-Guerrero et al., 2010)
	SBA-15	0.44	45	1	(Sanz et al., 2012)
	Silicalite	1.33	30	0.70	(Dunne et al., 1996)
Carbonaceous materials	Activated carbon (AC)	2.25	25	1	(Chen et al., 2012)
	Carbon nanotubes (CNTs)	1.70	20	1	(Hsu et al., 2010)
	CNTs modified by 3-aminopropyl-triethoxysilane (CNT(APTS))	2.61	20	1	(Hsu et al., 2010)
	Activated fly ash-derived carbon impregnated with MEA	1.56[h]	30	1	(Maroto-Valer et al., 2008)
	Wood ash modified with TEPA (45% wt.)[i]	2.02[j]	60	–	(Wang et al., 2017)
Metal-organic frameworks (MOFs)	Copper(II) benzene-1,3,5-tricarboxylate (CuBTC)	4.70	22	1	(Wang et al., 2002)
	IRMOF-1	1.10	25	1.2	(Millward and Yaghi, 2005)
	IRMOF-3	1.20	25	1.1	(Millward and Yaghi, 2005)
	IRMOF-6	1.10	25	1.2	(Millward and Yaghi, 2005)
	IRMOF-11	1.80	25	1.1	(Millward and Yaghi, 2005)
	MIL-100(Cr)	2.20	30	1	(Llewellyn et al., 2008)
	MIL-101(Cr)[k]	14.40	30	1	(Llewellyn et al., 2008)
	MOF-177	0.80	25	1	(Millward and Yaghi, 2005)
	MOF-2	0.60	25	1	(Millward and Yaghi, 2005)
	MOF-505	3.30	25	1.1	(Millward and Yaghi, 2005)

(*continued on next page*)

Table 7 (*continued*)

Adsorbent Type	Adsorbents	CO_2 Adsorption Capacity (mol CO_2/kg adsorbent)[a]	Operating Temperature (°C)	Operating Pressure (bar)	References
	MOF-74	4.90	25	1.1	(Millward and Yaghi, 2005)

Notes:
[a] Feed gas composition with pure CO_2, unless otherwise stated.
[b] Feed gas with 10% vol. CO_2 in N_2.
[c] Feed gas with 15% vol. CO_2 and 2.6% vol. H_2O vapour in N_2.
[d] Feed gas with 99.8% vol. CO_2.
[e] Feed gas with 99.9% vol. CO_2.
[f] Feed gas with 10% vol. CO_2 in argon (Ar).
[g] Synthesised by fusion method.
[h] Feed gas with 99.8% vol. CO_2.
[i] Ash collected from straw-fired power plants.
[j] Feed gas with 5% vol. CO_2 and 5% H_2O vapour in N_2.
[k] Activated by ethanol and ammonium fluoride (NH_4F) solution.

walled carbon nanotubes (CNTs), multi-walled CNTs, ordered mesoporous carbon and etc. Another approach is to increase the alkalinity by surface modification by incorporating additives or amines into the carbon structures (Wang et al., 2011). Hsu *et al.* (2010) showed that multi-walled CNTs modified by 3-aminopropyl-triethoxysilane (APTS) solutions (CNT(APTS)) could be cost-effective sorbents for CO_2 capture from flue gases as they are stable for prolonged adsorption-desorption cyclic operation (up to 20 cycles) with a maximum CO_2 adsorption capacity of 2.61 mol CO_2/kg adsorbent at 20 °C and 1 bar (Hsu et al., 2010).

Cheaper carbon resources such as fly ash and wood ash can also be used as CO_2 sorbents whilst reducing the environmental impacts of their disposal. However, their CO_2 adsorption capacities are relatively low and not practical for large-scale applications in CO_2 capture plants. Thus, current research focussed on improving the CO_2 adsorption capacity by incorporating amines into the sorbent frameworks. For example, Maroto-Valer *et al.* (2008) reported that activated fly ash-derived carbon with MEA modification exhibited higher CO_2 adsorption capacity (1.56 mol CO_2/kg adsorbent) than fly ash-derived carbon without activation (1.02 mol CO_2/kg adsorbent) due to combined physical adsorption inherent from the activated fly ash-derived carbon and chemical adsorption of the loaded MEA (Maroto-Valer et al., 2008). Recently, Wang *et al.* (2017) showed that wood ash-derived carbon with TEPA modification has superior capability for CO_2 capture and sorbent regeneration over other amine modifications on the parent sample by achieving a maximum CO_2 adsorption capacity of 2.02 mol CO_2/kg adsorbent with an amine loading of 45% wt. and maintaining satisfactory stability during 10 cyclic operations (Wang et al., 2017).

MOFs are increasingly considered as highly potential adsorbents for CO_2 capture and storage due to their high surface area, large pore volume and tunable pore surface properties. So far, MIL-101(Cr) activated by ethanol and ammonium fluoride (NH_4F) solution showed the highest CO_2 adsorption capacity of 14.40 mol CO_2/kg adsorbent at 1 bar and it can reach up to 40 mol CO_2/kg adsorbents (or 390 cm^3 CO_2 at standard temperature and pressure (STP)/cm^3 adsorbent) at 50 bar and 30 °C with pure CO_2 (Llewellyn et al., 2008). Most MOFs have low CO_2 adsorption capacity at low pressure and their adsorption capacities reduce significantly when exposed to gas mixtures. Their structure is sensitive to moisture, which affects their durability and mechanical strength. Additionally, the synthesis of MOFs is very expensive in many cases due to complicated synthesis process and low product yield. Due to these reasons, the use of MOFs in industrial applications is limited and more studies are required to develop economically viable MOFs with improved CO_2 capture performance and tolerance to contaminants such as H_2O vapour, SO_2 and NO_x (Lee and Park, 2015). One emerging technology is the fabrication of novel structured CO_2 adsorbents such as MIL-101(Cr) monoliths, which was 1.3 times more porous than 13X zeolite monoliths, thermally regenerable and have the capability to adsorb high concentration of CO_2 at 40% vol. in the case of biogas upgrading (Hong et al., 2020).

Traditionally, adsorbents are regenerated by pressure swing adsorption (PSA) or temperature swing adsorption (TSA). However, PSA process has intensive energy requirements while the TSA process has a long regeneration time, which will increase the cost of regeneration and reduce the desorption efficiency (Yuan and Eden, 2015). Therefore, several new and more efficient adsorbent regeneration processes have been developed to improve the desorption efficiency and reduce the energy requirement for regeneration. Grande *et al.* (2009) found that CO_2 capture from NGCC power plants with electric swing adsorption (ESA) has a short regeneration time (<3 min) although the process gave low CO_2 recovery of 80% vol. with a purity of 80% vol., and requires an energy consumption of 2.04 GJ/t CO_2 to heat a 13X zeolite honeycomb monolith electrically (Grande et al., 2009). Thili *et al.* (2009) reported that vacuum swing adsorption (VSA), in which the desorption step is performed under vacuum only, would lead to pure CO_2 with purity around 99% but limited recovery of 85% vol. CO_2 (Tlili et al., 2009). VSA has also been known to have higher efficiency over PSA as there is no requirement to pre-compress the flue gas to high pressure (Zanco et al., 2021).

Plaza et al. (2010) compared different regeneration methods (i.e., TSA, VSA and vacuum and temperature swing adsorption (VTSA)) for post-combustion CO_2 capture with commercial activated carbon and they found that the productivity and CO_2 recovery followed the order: TSA < VSA < VTSA (Plaza et al., 2010). A maximum productivity of 1.9 mol/kg/h and a CO_2 recovery up to 97% vol. was achieved by VTSA operation. Furthermore, Jiang *et al.* (2020) found that a two-stage vacuum and pressure swing adsorption (VPSA) process consisting of 8-steps with 13X zeolite adsorbent is the most effective process for CO_2 capture from flue gas because it has the lowest energy consumption (0.79 GJ/t CO_2) compared to the temperature and vacuum swing adsorption (TVSA) process (3.22 GJ/t CO_2) and the TSA process (6.76 GJ/t CO_2) (Jiang et al., 2020). They also found that TVSA gives the best separation performance with CO_2 purity of 92% vol. and CO_2 recovery of 98% vol. than VPSA (CO_2 purity of 96% vol. and CO_2 recovery of 92% vol.) and TSA (CO_2 purity of 95% vol. and CO_2 recovery of 90% vol.).

3.2.3. Membrane

Membrane separation can be used to selectively separate CO_2 from the flue gas through a permeable/semipermeable material by chemical/physical mechanisms such as solution-diffusion transport, molecular sieving, surface diffusion and Knudsen diffusion (Yuan and Eden, 2015). Membrane separation is a pressure- and/or concentration-driven process, hence it is more favourable and less energy-intensive for pre-combustion processes (due to high CO_2 partial pressure and/or concen-

Table 8
CO_2 permeability, CO_2/N_2 selectivity and operating conditions of different polymeric membrane materials (Songolzadeh et al., 2014).

Material	CO_2 Permeability (barrer)[a]	CO_2/N_2 Selectivity (–)	Feed Pressure (bar)	Temperature (°C)
Copolymers and polymer blends	0.8 – 6.1 × 10^5	3.2 – 58.0	1 – 10	0 – 100
Polyacetylenes	15 – 19,000	5.0 – 23.0	–	25
Polyarylates	1.2 – 85.1	14.7 – 29.1	10	35
Polyarylene ethers	18.5 – 29.5	11.6 – 13.5	1	35
Polycarbonates	2.2 – 18.6	15.0 – 26.3	1 – 10	35
Polyethylene oxide	8.1 – 773	44.0 – 140.0	4.4 – 14.6	25 – 45
Polyimides	0.5 – 600	16.3 – 38.5	0.4 – 10	25 – 35
Polyphenylene oxides	1.6 – 159.9	18.7 – 34.8	1.5 – 6.9	22 – 35
Polypyrroles	0.1 – 54	20.8 – 46.3	10	35
Polysulfones	1.4 – 105	9.0 – 32.0	1 – 35	35

tration in the flue gas streams) than post-combustion processes (due to low CO_2 partial pressure and/or concentration in the flue gas streams) (Wang and Song, 2020; Zhao et al., 2010). There are no moving parts in membrane separation systems and the membrane modules can be easily installed into existing infrastructures (Yuan and Eden, 2015). However, a high degree of CO_2 removal with high purity (>95% vol.) cannot be achieved by a single-stage membrane process due to the low practical transmembrane pressure ratio and membrane selectivity, therefore a multi-stage membrane process has to be designed (IEA, 2021d; Zhao et al., 2010). Sophisticated recycling streams can also be designed in the membrane processes to enhance the CO_2 flux to achieve higher CO_2 recovery at 90% vol. (Han et al., 2020). Zanco *et al.* (2021) analysed that membrane- and adsorption-based processes may become cost competitive to absorption-based process for small scale post-combustion CO_2 capture plant that processed less than 100 tonnes of flue gas in a day with recovery rates below 40% vol. CO_2 (Zanco et al., 2021). Despite the existing challenges in applying membranes to post-combustion processes, this technology has been commercially employed for removing CO_2 from natural gas streams (Yuan and Eden, 2015).

Various types of membranes have been developed for CO_2 capture, namely: inorganic membranes, polymeric membranes, mixed matrix membranes (MMMs), facilitated transport membranes (FTMs) and gas-liquid membrane contactors (Khalilpour et al., 2015; Belaissaoui and Favre, 2014). Inorganic membranes are those that made up of zeolites, oxides (such as Al_2O_3, titanium dioxide (TiO_2), zirconium dioxide (ZrO_2)), carbons or MOFs as top membrane layer that is casted on a porous support (usually metal or ceramic) for mechanical stability. Inorganic membranes have high thermal stability and good mechanical stability but their low CO_2 permeability and high fabrication cost limits their scale-up (Al-Mamoori et al., 2017; Spigarelli and Kawatra, 2013). Thus, current membrane-based commercial applications used polymeric membranes due to their ease of manufacture, low production cost, excellent CO_2 separation performance and good mechanical stability (Songolzadeh et al., 2014). A disadvantage of polymeric membranes is that they have very low thermal stability, which limits their application in post-combustion CO_2 capture. The temperature of the flue gases have to be cooled down prior to the membrane separation process to reach a higher degree of selectivity and consume less energy (Zhao et al., 2017).

A wide variety of polymeric membrane materials have been developed and their CO_2 permeability, CO_2/N_2 selectivity and operating conditions are summarised in Table 8. The most popular polymeric membranes for CO_2 separation are made of polyarylates, polycarbonates, polyimides, and polysulfones (Spigarelli and Kawatra, 2013). A new type of polymeric membranes is polymers of intrinsic microporosity (PIM) membranes, which are promising candidates for CO_2 capture because they show high CO_2 permeability and selectivity. The PIM hollow fibre membranes fabricated by Jue *et al.* (2017) exhibited a relatively high CO_2 permeability of 1,008 barrer with high CO_2/CH_4 selectivity of 22.5 and high CO_2/N_2 selectivity of 27.7 but it has a low CO_2/H_2 selectivity of 1.0 (Jue et al., 2017).

MMMs are another type of promising materials for CO_2 capture due to their enhanced CO_2 permeability, selectivity, and thermal and mechanical stabilities. MMMs have a composite structure, consisting of polymers (such as polysulfones, polycarbonates, polyarylates, poly(arylketones), poly(arylethers), polyimides or block copolymers) and inorganic fillers (such as zeolites, carbon nanotubes, silicates, alumina, metal oxides or MOFs) (Yuan and Eden, 2015). A key challenge in the fabrication of MMMs is to ensure good adhesion between the polymer and inorganic particles to prevent void formations, which may reduce their selectivity (Khalilpour et al., 2015). Ghalei *et al.* (2017) reported that the selectivity of MMMs for CO_2 capture could be enhanced through efficient dispersion of amine functionalised MOF nanoparticles to minimize non-selective microvoid formation around the particles (Ghalei et al., 2017). Shahid *et al.* (2014) showed that increased loading of mesoporous iron 1,3,5-benzenetricarboxylate (Fe-BTC) nanoparticles, an inorganic material, in polymeric MMMs-based on Matrimid®-polyimide (PI) would improve the CO_2 permeability and CO_2/CH_4 selectivity by 62% and 30% compared to the native Matrimid®-PI membrane at 40 bar as well as reduce the tendency of plasticization at high pressures (Shahid and Nijmeijer, 2014). Despite the great potentials shown by MMMs over polymeric and inorganic membranes for CO_2 capture, their fabrication is costly and they are not yet practical for industrial applications (Al-Mamoori et al., 2017).

FTMs such as liquid-supported membranes, ion-exchange membranes and fixed-carrier membranes are promising candidates as next-generation membrane for separating CO_2 from high- and low-pressure gas streams. The incorporation of functional groups on the polymer matrix makes them exhibiting high CO_2 permeability, high CO_2/N_2 and CO_2/H_2 selectivities, and superior material stability and mechanical properties (Yuan and Eden, 2015). For example, a relatively high CO_2 permeability between 2,700 barrer and 3,100 barrer, and a high CO_2/N_2 selectivity between 30 and 33 at pressure ranging from 0.2 bar to 1 bar were reported for liquid-supported membrane immobilized with 1-ethyl-3-methylimidazolium-bis(trifluoromethylsulfonyl) imide [emim][Tf_2N], an IL, in polyvinylidene fluoride (PVDF) hollow fibre support (Kim et al., 2011). A higher CO_2/N_2 separation performance was observed using a polyvinylamine (PVAm) modified with a cross-linking agent containing carriers piperazine (PIP) on polysulfone (PS) ultrafiltration (PVAm–PIP/PS) composite membrane with CO_2 permeability of 195,000 barrer and CO_2/N_2 selectivity of 277 at 22°C and 1.1 bar (Qiao et al., 2012). FTMs are currently under pilot-scale test and they still have fabrication challenges to produce defectless membranes and to use nanosized carriers without agglomeration (Wang and Song, 2020). He *et al.* (2015) estimated that CO_2 capture from flue gas using fixed-carrier membrane has an energy consumption of 1.02 GJ/t CO_2 with a CO_2 capture cost of USD 47.87/t CO_2 (He et al., 2015).

Gas-liquid membrane contactors are highly effective hybrid separation systems that use a porous, hydrophobic polymeric membrane as an interface between a flue gas stream and an absorption liquid (such as amine, ILs and etc.) that flow counter-currently instead of column configurations (such as packed beds, spray towers and etc.)

(Khalilpour et al., 2015). CO_2 will diffuse through the membrane and absorb by the absorption liquid. The remaining flue gas is released to the atmosphere (Spigarelli and Kawatra, 2013). The advantages of gas-liquid membrane contactors over other types of membranes are no requirement to pressurize the flue gas (which reduces the energy consumption), higher CO_2 fluxes, no selective layer, independent flow regulation and lower mass transfer resistance from the membrane when compared to the gas and liquid mass transfer resistance (Rivero et al., 2020). There are several configurations of membrane modules, which include tubular, flat sheet/plate-and-frame, spiral wound and hollow fibre. The most popular types of modules for industrial CO_2 separation is hollow fibre membranes because they exhibit high production rates due to their optimum geometry, high surface-to-volume ratio and more compact than flat sheet or spiral wound units (Al-Mamoori et al., 2017).

Cao *et al.* (2021) reported that the efficiency of CO_2 removal for MDEA-based nanofluid in a hollow fibre membrane contactor with 0.5% wt. CNT nanoparticles was improved by about 8% (on average) when the MDEA concentration was increased from 5% wt. to 20% wt. and the solvent flow rate was varied from 10 ml/min to 40 ml/min (Cao et al., 2021). They also found that the CO_2 removal efficiency of the module was increased by 14% as the membrane porosity increased from 0.2 to 0.7 and decreased by about 30% as the membrane tortuosity increased from 1 to 25. Simons *et al.* (2010) showed that the potential use of amino acid salt solution (sarcosine) as a competitive absorption liquid to MEA in a membrane contactor for CO_2/CH_4 separation (Simons et al., 2010). Their study revealed that a membrane contactor with sarcosine could yield significantly higher CO_2/CH_4 selectivity (over 70 and it could almost reach 120 when the temperature difference between absorber and desorber are 0 °C and 35 °C, respectively) compared to the conventional MEA (CO_2/CH_4 selectivity over 20 and 65 under the same conditions) although they exhibit lower CO_2 permeance than MEA. Additionally, significantly higher CH_4 product yields could be obtained as CH_4 loss is reduced due to low absorption of CH_4 in sarcosine compared to MEA.

3.2.4. Cryogenic

Cryogenic technology involves separating CO_2 from other components present in the flue gas through several stages of compression and cooling at very low temperatures (–100 °C to –135 °C) and high pressures (101 bar to 203 bar) to produce liquid CO_2 of high purity (99.99% vol.) that is ready for transportation and storage (Leung et al., 2014, Song et al., 2019). This separation technique can be applied to pre-combustion or oxy-fuel combustion CO_2 capture systems (Mondal et al., 2012). The advantages of cryogenic CO_2 separation are that no chemical absorbents are required, an extremely high CO_2 recovery of 99.99% can be obtained and the process can be operated at atmospheric pressure. However, there are some challenges with this cryogenic separation, which are the high possibility of process blockage (due to formation of ice in the CO_2 purification unit and/or formation of solid CO_2 on the heat exchanger surface), high energy requirement for regeneration (due to extremely low temperature and high pressure employed in the process) and high CO_2 capture cost (due to large pressure drop during operation and several costly steps to remove H_2O entirely from the flue gas) (Tuinier et al., 2010).

Several types of cryogenic CO_2 capture processes have been developed to improve the technical performance of this technology, which include packed bed, external cooling loop, anti-sublimation, distillation, controlled freeze zone, CryoCell and Stirling cooler systems. Their CO_2 capture performance and energy consumption are provided in Table 9. Tuinier *et al.* (2011) showed that cryogenic CO_2 capture using dynamically operated packed beds at a 600 MW coal-fired power plant was cost competitive (higher CO_2 avoidance cost at USD 126.50/t CO_2 avoided) to amine scrubbing (USD 54.50/t CO_2 avoided) and membrane (USD 120/t CO_2 avoided) processes when the cold energy is provided by low-cost liquefied natural gas (LNG) (Tuinier et al., 2011). Additionally, this cryogenic packed bed process can simultaneously separate H_2O and CO_2 from the flue gas based on the differences in their dew and sublimation points, which will prevent clogging and large pressure drops during operation. An effective way to reduce energy consumption of cryogenic CO_2 capture is to reuse waste cold energy from industrial sources such as LNG via an external cooling loop. Jensen *et al.* (2015) revealed that the external cooling loop cryogenic CO_2 capture process for a 550 MW coal-fired power plant could achieve an extremely high purity of liquid CO_2 at 99.2% vol. and a low energy consumption of 0.74 GJ/t CO_2 using an internal carbon tetrafluoride (CF_4) refrigeration cycle to transfer heat from melting CO_2 to desublimating CO_2 by cooling the contact liquid and an external cooling loop of natural gas or other refrigerant to provide additional heat duty to operate the cryogenic process (Jensen et al., 2015). Anti-sublimation CO_2 capture is another type of cryogenic process based on the concept of frosting and defrosting CO_2 at atmospheric pressure in a low temperature evaporator. Clodic *et al.* (2005) estimated that the anti-sublimation CO_2 capture from flue gas at a conventional pulverized coal-fired power boiler has a low energy consumption of 1.25 GJ/t CO_2 for a 90% CO_2 removal with about 21% lower energy penalty than MEA-based CO_2 capture (Clodic et al., 2005).

Cryogenic distillation is widely used for CO_2 separation from gas streams containing high CO_2 concentration (>50% vol.) at high pressures (Mondal et al., 2012). Xu *et al.* (2014) proposed an improved cryogenic distillation process for CO_2 capture and their study showed that the process could achieve a very high CO_2 purity of 99.9% vol. with an extremely low energy consumption of 0.43 GJ/t CO_2 at a lower CO_2 capture cost (USD 10.28/t CO_2) compared to absorption process using MEA (USD 24/t CO_2) and Selexol (USD 19/t CO_2) for post-combustion CO_2 capture (Xu et al., 2014). They also estimated that the energy consumption would increase to 0.97 GJ/t CO_2 and the CO_2 capture cost would increase to USD 18.46/t CO_2 when cryogenic distillation was employed in oxy-fuel combustion CO_2 capture system since a supply of O_2 is needed for oxy-fuel combustion. Controlled freezing zone is a cryogenic single distillation column technology introduced by ExxonMobil to remove CO_2 and impurities such as hydrogen sulfide (H_2S) from natural gas. This technology has been successfully demonstrated at Clear Lake pilot plant and commercially demonstrated at Shute Creek gas treating facility in LaBarge, Wyoming in the US, showing high CO_2 recovery between 99.5% vol. to 100% vol. CO_2 at 41 bar for a feed gas stream of high CO_2 concentration (i.e., 42% vol. and 71% vol.) (Condon, 2012).

Another cryogenic process for CO_2 separation from natural gas is CryoCell technology, in which the separation is based on CO_2 solidification property. This technology was developed by Cool Energy Ltd. and tested in collaboration with other industrial partners, including Shell Global Solutions, in a demonstration plant in Western Australia. The advantages of CryoCell technology include no requirement to supply and treat process makeup water, no process heating system is required, no chemicals usage, no corrosion as water is removed immediately downstream of the inlet separator and no foaming potential. Hart *et al.* (2009) showed that CryoCell technology has lower total energy requirement and total plant costs (by 81% and 33%, respectively) compared to amine technology, despite having a low CO_2 removal efficiency (up to 34% vol. with feed gas pressure between 55 bar and 65 bar) (Hart and Gnanendran, 2009). A Stirling cooler system for cryogenic CO_2 capture from post-combustion flue gas was designed by Song et al. (2012). The main advantage of the system is that it can be operated at atmospheric pressure and solvent regeneration and energy penalty of pressure drop can be avoided. They showed that the Stirling cooler system can achieve 85% vol. CO_2 removal from the flue gas with an energy consumption of 3.40 GJ/t CO_2, which is comparable to a membrane-based CO_2 capture process (3.51 GJ/t CO_2).

3.2.5. Biological

Biological CO_2 capture involves plants and micro-organisms (such as bacteria, algae, fungi and yeast) for removing CO_2 from flue gas and the atmosphere. Common biological methods are forestation, ocean fertilisation and biophotolysis using cyanobacteria or microalgae (Yang et al., 2008). CO_2 emissions in the atmosphere can be reduced by different

Table 9
CO_2 capture performance and energy consumption of different cryogenic separation processes.

Cryogenic Process	Flue Gas Composition	Cold Energy Source	CO_2 Recovery (% vol.)	CO_2 Purity (% vol.)	Energy consumption (GJ/t CO_2)	References
Packed bed	10% vol. CO_2 and 1% vol. H_2O in N_2	Liquidfied natural gas (LNG)	99.5	–	1.80	(Tuinier et al., 2011)
External cooling loop	14% vol. CO_2, 2% vol. O_2, 15% vol. H_2O, 1% vol. Ar in N_2	Carbon tetrafluoride (CF_4) refrigerant and LNG	93.3	99.2	0.74	(Jensen et al., 2015)
Anti-sublimation	10% vol. CO_2, 6% vol. O_2, 14% vol. H_2O in N_2	Mixed refrigerant	90.0	–	1.25	(Clodic et al., 2005)
Distillation	80% vol. CO_2, 5% vol. O_2, 5% vol. Ar in N_2	Compressor and heat exchanger (cooler)	90.0	99.9	0.43	(Xu et al., 2014)
Controlled freezing zone	42% vol. to 71% vol. CO_2	Refrigerant	99.5 to 100	–	–	(Condon, 2012)
CryoCell	20% vol. to 35% vol. CO_2	Chiller	34.0	–	–	(Hart and Gnanendran, 2009)
Stirling cooler	13% vol. CO_2 in N_2	Stirling cooler	85.0	–	3.40	(Song et al., 2012)

Table 10
Comparison of different forestation options for atmospheric CO_2 capture (Smith et al., 2014).

Options	CO_2 Capture Potential per Area (t CO_2/ha)	Ease of Implementation	Implementation Timescale (Years)
Reducing deforestation	1 – 10	Medium	Immediate
Afforestation/reforestation	1 – 10	Easy	Immediate
Forest management	1 – 10	Medium	Immediate
Forest restoration	1 – 10	Medium	Immediate

forestation options such as reducing deforestation to conserve existing CO_2 pools in forest vegetation and soil, afforestation/reforestation to improve biomass stocks, forest management for sustainable timber production and forest restoration to protect secondary forests and other degraded forests (Smith et al., 2014). All these forestation options have a medium CO_2 capture potential per area (1 t CO_2/ha to 10 t CO_2/ha) and they can be implemented immediately (Table 10). Among the forestation options, afforestation/reforestation is the easiest to implement, for example, by planting trees on non-forested agricultural lands. Fuss *et al.* (2018) estimated that afforestation/reforestation could potentially remove 0.5 Gt CO_2/year to 3.6 Gt CO_2/year in 2050 at a cost of USD 5/t CO_2 to USD 50/t CO_2 (Fuss et al., 2018).

Ocean fertilisation is another biological method for capturing atmospheric CO_2 by phytoplanktonic photosynthesis process (Nogia et al., 2016). Ocean fertilisation can be achieved by increasing the supply of limiting nutrients (such as nitrogen, phosphorus and iron) to the near/sunlit-surface ocean or by accelerating nutrient re-supply from mid-deep waters (also known as mesopelagic layer or twilight zone) to simulate the growth of marine phytoplankton (free-living microscopic marine plants) and potentially enhance their production at all tropic levels. However, it is extremely challenging to quantify with acceptable accuracy the CO_2 removed from the atmosphere by large-scale ocean fertilisation on long-term and to sufficiently monitor unintended impacts (for example, on the marine ecosystem) over large space and time scales (Williamson et al., 2012).

In biophotolysis process, micro-organisms cultivated in a photoreactor convert sunlight (light energy), H_2O and CO_2 under anaerobic condition by photosynthesis into useful products such as carbohydrates, H_2 and O_2 (Equation 9 and Equation 10). Sometimes, this process is also called water-splitting photosynthesis. The light energy absorbed by photoautotrophic organisms (such as cyanobacteria and microalgae) is stored as chemical energy in the form of carbohydrates in the organisms' cells. The production of H_2 by microalgae via a water photolysis reaction is catalysed by the hydrogenase enzyme (Akkerman et al., 2002). Jacob-Lopes *et al.* (2008) showed that biological CO_2 capture by Aphanothece microscopica Nägeli, a cyanobacterium, was affected by the CO_2 concentration, temperature and light intensity in the photoreactor (Jacob-Lopes et al., 2008). They found that this bacterium could achieve a maximum specific growth rate of 0.04/h and a high CO_2 removal rate of 109.2 mg/L/h at 15% vol. CO_2, 35 °C and 11 klux.

$$6CO_2 + 6H_2O + \text{light energy} \rightarrow C_6H_{12}O_6 + 6O_2 \quad (9)$$

$$4H_2O + \text{light energy} \rightarrow 2O_2 + 4H_2 \quad (10)$$

Although biophotolysis is the most desirable and attractive H_2 production process due to its low cost, the main issue with this process is that O_2 is also generated during photosynthesis since the hydrogenase enzyme is strongly inhibited by O_2 and that may limit the process efficiency. Other drawbacks include requirement for customised photobioreactors and low H_2 yield due to very low efficiency of light conversion. Several approaches have been suggested to improve the process efficiency, which include development of O_2-tolerant hydrogenases, replacement of the hydrogenase with nitrogenase in cyanobacteria, use an indirect biophotolysis process that separate photosynthetic water splitting and H_2 evolving reactions in time or place, and changes in operating conditions. To date, direct biophotolysis process is still in laboratory experimentation stage (Chandrasekhar et al., 2015).

On the other hand, microalgae-based CO_2 capture can absorb capture CO_2 from flue gases at power plants and from the atmosphere, and convert it into organic matter by photosynthesis to produce lipids, proteins and carbohydrates. The generated biomass can be used as a feedstock for biofuel, functional food and animal feed (Hosseini et al., 2018). Microalgae-based CO_2 capture has been widely studied due to their high photosynthetic capability and fast growth rate. However, the main challenges of microalgae-based CO_2 capture are low CO_2 capture/fixation efficiency for open-culture systems (typically between 10% vol. and 40% vol.), low CO_2 solubility, high cost of CO_2 capture and transport, significant CO_2 loss during algae culture and weak tolerance to high CO_2 concentration for most of the microalgae as they typically grow at low CO_2 concentration (below 5% vol.) (Song et al., 2019; Chi et al., 2011; Zhao and Su, 2014). Some microalgae can grow at flue gas concentration (between 10% vol. and 15% vol.) and very few microalgae can tolerate high CO_2 concentration (for example, *Chlorella* sp. KR-1 and *Chlorella*

ZY-1 can tolerate up to 70% vol. CO_2 and *Chlorella* sp. T-1 can tolerate up to 100% vol. CO_2) (Zhao and Su, 2014).

Closed photobioreactors can be used to reduce CO_2 losses since they can prolong CO_2 retention time and enhance the CO_2 gas-liquid mass transfer efficiency. Li *et al.* (2013) reported that the CO_2 fixation efficiency by microalgae (*Chlorella vulgaris*) in a closed raceway pond was increased to 95% vol. under intermittent aeration (Li et al., 2013). Another issue with biological CO_2 capture using microalgae is its extremely high energy requirement for microalgae cultivation, particularly for closed flat plate and tubular photobioreactors (8.31 GJ/t CO_2 and 70.36 GJ/t CO_2, respectively). Jacob *et al.* (2015) reported that CO_2 capture by microalgae in closed flat plate and tubular photobioreactors was 2.2 and 19 times more energy intensive, respectively, than CO_2 capture by MEA (3.80 GJ/t CO_2) while CO_2 capture by microalgae in open raceway pond is 0.8 times less energy intensive (corresponds to 3.09 GJ/t CO_2) than CO_2 capture by MEA (Jacob et al., 2015). Moreover, the net energy ratio (i.e., the ratio of energy output in gaseous biofuel to energy input to the microalgae cultivation system for pumping and circulation) for CO_2 capture by microalgae in open raceway pond system was higher (0.71) when compared to those in closed flat plate and tubular photobioreactors (0.42 and 0.05, respectively). The CO_2 capture cost using microalgae cultivated in open ponds was estimated at USD 793/t CO_2, excluding the costs of transportation and burial of produced biomass (Farrelly et al., 2013). This value was cost intensive when compared with other CO_2 separation technologies (Table 3). Further process optimisation and improvement on the photoreactor system are required to develop a technically and economically viable microalgae-based CO_2 capture process for industrial applications.

3.3. CO_2 transport

CO_2 transport is an important part of the CCS chain that link CO_2 sources to storage sites via pipelines, ships, trucks or rails. Large amount of captured CO_2 can be transported by compressing the CO_2 to dense phase (i.e., in supercritical form at pressure and temperature above 74 bar and 31 °C, respectively) for pipeline transport or by refrigerating the CO_2 to liquid phase for ship, truck or rail transport. Normally, H_2O is present in the captured CO_2 and it must be removed before transportation to prevent the acid formation that can corrode pipelines and other equipment. Dehydration of the captured CO_2 gas stream is typically performed together with compression or refrigeration. The indicative costs of CO_2 compression and dehydration for a CCS project at US Gulf coast in 2020 was between USD 12.76/t CO_2 for compressing and dehydrating 5 Mt CO_2/year and USD 23.45/t CO_2 for compressing and dehydrating 400 kt CO_2/year with captured CO_2 gas stream containing <200 ppmv H_2O at 150 bar (Kearns et al., 2021; World Resources Institute (WRI), 2008).

3.3.1. Pipeline transportation

So far, all current and proposed large-scale transport of CO_2 is by pipelines since it was a mature technology (TRL 9). It was reported that more than 50 Mt CO_2/year could be transported by pipelines over long distances on land (>2,500 km) as well as in the seas and oceans (up to 2.2 km deep) from CO_2 sources, particularly power plants with a lifetime longer than 23 years, to EOR operations (National Petroleum Council, 2019a; Norişor et al., 2012). Commercial CO_2 pipelines typically operate at pressures between 86 bar and 152 bar (to avoid two-phase flow regimes and increase the density of CO_2, thus making it easier and less costly to transport), and temperatures between 13 °C and 43 °C (to avoid damage on the external pipe coating and effect on the pipelines' integrity at high temperatures, and to avoid effect on the metal used to construct the pipelines at very low temperatures) (Metz et al., 2005; World Resources Institute (WRI), 2008; National Petroleum Council, 2019a).

The transportation costs vary with the modes, capacity and distance of transportation. Pipeline transport can be onshore or offshore, depending on the source-to-storage location. Offshore pipeline transport is often more costly than onshore pipeline transport. Zero Emissions Platform (ZEP) (2011) reported that the cost of offshore pipeline transport for a single CCS demonstration project of 2.5 Mt CO_2/year in Northern Europe was USD 10.69/t CO_2 for every 180 km, which was 1.7 times more than the cost of onshore pipeline transport (USD 6.21/t CO_2) with the same capacity and distance (Table 11) (ZEP, 2011). Additionally, the cost of CO_2 transportation was increased by 5.6 times (from USD 10.69/t CO_2 to USD 59.43/t CO_2) when the offshore pipeline transport distance was increased from 180 km to 1,500 km for a single CCS demonstration project of 2.5 Mt CO_2/year in Northern Europe. The pipeline transport costs vary from country to country. For example, it was reported that the cost of CO_2 onshore pipeline transport for a CCS project at US Gulf coast in 2020 was between USD 2.41/t CO_2/180 km for transporting 20 Mt CO_2/year and USD 24.48/t CO_2/300 km for transporting 1 Mt CO_2/year (Kearns et al., 2021).

Although pipeline transport provides significant economies of scale for high volumes and flow rates of CO_2, pipelines are capital intensive investments (National Petroleum Council, 2019a). One way to reduce the cost of CO_2 pipeline transport is to implement large-scale CCS networks. Table 11 shows that the costs of CO_2 onshore pipeline transport could be lower by 3.6 times for every 180 km and the costs of CO_2 offshore pipeline transport could be lower by 2.7 times for every 180 km and 3.2 times for every 1,500 km for large-scale CCS networks of 20 Mt CO_2/year with double feeders and double distribution pipelines when compared with a single CCS demonstration project of 2.5 Mt CO_2/year. Offshore networks that combine pipeline and ship transportation could offer a cost-effective solution, particularly for CCS clusters in their early development (ZEP, 2011).

3.3.2. Ship transportation

CO_2 transport by ships is based on the shipping experience in the food and beverage industries and it a mature technology (TRL 9) as it has been practised for over 30 years at small-scale, with only 3 Mt CO_2/year. CO_2 shipping is now considered for large-scale transport of CO_2 because it may be more economical when CO_2 needs to be transported on a large-scale over large distances or overseas than constructing new long-distance pipelines or repurposing gas pipelines at existing loading facilities and unloading platform (World Resources Institute (WRI), 2008). The long commercial experience of the gas industry with over 80 years in shipping various pressurized gases could be adapted to CO_2 shipping with similar port infrastructure as those for LNG and liquified petroleum gas (LPG). It was estimated that an average ship/tanker can carry about 45 kt liquified CO_2 at 17 bar and –40 °C (Kearns et al., 2021; National Petroleum Council, 2019a).

Shipping has different economies of scale to pipelines and it can be deployed in one ship or more, which can be directed to different storage sites. Unlike pipeline transport, ship transport is less capital intensive and cost competitive for trans-oceanic movements (National Petroleum Council, 2019a). ZEP (2011) reported that the CO_2 shipping costs for a CCS project in northern Europe was between USD 12.76/t CO_2/180 km for shipping 20 Mt CO_2/year and USD 16.67/t CO_2/1,500 km for shipping 2.5 Mt CO_2/year (Table 11) (ZEP, 2011). Meanwhile, the cost of CO_2 shipping transport for a CCS project at US Gulf coast in 2020 was between USD 14.48/t CO_2/180 km for shipping 20 Mt CO_2/year and USD 23.45/t CO_2/1,500 km for shipping 2.5 Mt CO_2/year (Kearns et al., 2021).

3.3.3. Truck and rail transportation

CO_2 transport by trucks (TRLs 8 to 9) and rails (TRLs 7 to 9) are viable methods for small quantities of CO_2 (from 4 tonnes to a few hundred tonnes) over shorter distances (<322 km for trucks and <1,609 km for rails) but they are not economical and scalable options when transporting large volumes of CO_2 in long-term compared to pipelines and ships (National Petroleum Council, 2019a). The typical pressure and temperature of CO_2 transported by trucks and rail tankers are –20

Table 11
Indicative costs of CO_2 transport by onshore and offshore pipelines, and ship for a single CCS demonstration project of 2.5 Mt CO_2/year and for large-scale CCS networks of 20 Mt CO_2/year in Northern Europe (ZEP, 2011).

Transport Modes	Single CCS Demonstration Project of 2.5 Mt CO_2/year		Large-Scale CCS Networks of 20 Mt CO_2/year	
	Transport Cost (USD/t CO_2/180 km)	Transport Cost (USD/t CO_2/1,500 km)	Transport Cost (USD/t CO_2/180 km)	Transport Cost (USD/t CO_2/1,500 km)
Onshore Pipeline	6.21	–	1.72	–
Offshore Pipeline	10.69	59.43	3.91	18.74
Ship	9.43	16.67	12.76	18.51

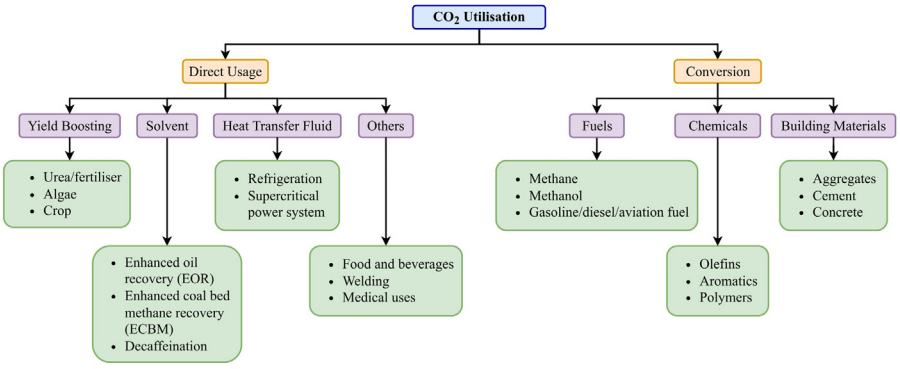

Figure 7. CO_2 utilisation pathways (IEA 2019).

°C and 20 bar (Metz et al., 2005). Trucks can complement ship transport to move small quantities of CO_2 (<10 kt) from port CO_2 terminals to industrial sites for utilisation and from CCS project sites to a nearby storage location for EOR that are inaccessible to pipeline options (Orchard et al., 2021). The cost of liquid CO_2 transported by truck is expected to increase when the sourcing radius was more than 241 km (National Petroleum Council, 2019a).

3.4. CO_2 utilisation

The captured CO_2 can be transported to receivers for utilisation, in which CO_2 is converted into a wide variety of commercially viable end-uses/products and for enhanced hydrocarbon recovery that can potentially offset the costs associated with CCS (Hepburn et al., 2019). IEA (2019) estimated that about 230 Mt CO_2/year are utilised globally with 57% of the global CO_2 demand is in the fertiliser industry for urea production, 34% in the oil sector for EOR, 3% for food production, 3% for beverages production, 2% for metal fabrication and 4% in other commercial applications such as cooling, fire suppression and crop cultivation in greenhouses (IEA, 2019). CO_2 transformation into fuels, chemicals and building materials are new pathways of CO_2 utilisation but most of them are highly energy-intensive and still in the research and development stages due to commercial and regulatory challenges.

CO_2 utilisation may not reduce CO_2 emissions and it may not deliver a net climate benefit because the amount of CO_2 utilised by a pathway is likely to differ from the amount of CO_2 captured or stored, with storage timeframes ranging from days to millennia (Hepburn et al., 2019). The quantification of climate benefits of CO_2 utilisation applications and their emission reduction potential is a complex process and it requires a comprehensive life-cycle assessment and understanding of its market dynamics. CO_2 utilisation is a complement but it is not an alternative to CO_2 storage for large-scale emissions reductions (IEA, 2019). Mac Dowell *et al.* (2017) estimated that the contribution of carbon capture and utilisation (CCU) to the global CO_2 mitigation challenge would be negligible (0.2 Gt CO_2/year in 2050) and it could not compete with CCS as it has a much higher CO_2 capture potential, which was estimated at 7.8 Gt CO_2/year in 2050 (Mac Dowell et al., 2017). There are a wide range of CO_2 utilisation applications, which involve direct usage of CO_2 or usage through conversion into other products (Figure 7). The estimated market and TRL of CO_2 utilisation are shown in Table 11.

3.4.1. Direct CO_2 utilisation

CO_2 can be used directly as: (i) a feedstock for urea yield boosting, algae cultivation and crop cultivation in greenhouses, (ii) a solvent in EOR, enhanced coal bed methane recovery (ECBM), decaffeination and dry cleaning, (iii) a heat transfer fluid in refrigeration and supercritical power system, and (iv) other applications such as food and beverages, welding and medical uses. This section will cover on the direct use of CO_2 for yield boosting in urea production, algae cultivation and crop cultivation in greenhouses. CO_2 utilisation for EOR and ECBM will be covered later in Section 3.5.

Urea ($CO(NH_2)_2$), a nitrogen fertiliser, is commercially produced by reacting high purity CO_2 with ammonia (NH_3) at high temperature and pressure (Equation 11). Typically, about 0.735 tonne of CO_2 to 0.75 tonne of CO_2 is utilised to produce 1 tonne of urea. The cost of urea is between USD 205/t urea to USD 285/t urea. When urea was applied to the land and contacted with H_2O, the NH_3 produced is absorbed by the plant and the resultant CO_2 is released to the atmosphere (Global CCS Institute, 2011). Urea yield boosting is a widely implemented and mature technology (TRL 9) (Patricio et al., 2017). The estimated global market for urea in 2016 is 180 Mt/year, utilising 132 Mt CO_2/year (Aresta et al., 2013). The main barriers of deploying urea yield boosting technology are the volatility in price and demand for urea and NH_3 makes long-term appraisal difficult and the potential high capital costs of CO_2 capture infrastructure (Global CCS Institute, 2011).

$$2NH_3 + CO_2 \rightleftharpoons CO(NH_2)_2 + H_2O \quad (11)$$

Algae cultivation to produce commercial petroleum substitutes is a novel application of CO_2. The production yields of algae can be greatly increase by bubbling dilute CO_2 flue gas through the algal cultivation systems. Typically, about 1.8 t CO_2 is utilised to produce 1 t dry algal biomass, which may vary with algae species. Algae cultivation pathway has complex production economics and the cultivation of 1 Mt/year of algae for biodiesel production was estimated to utilise 2 Mt CO_2/year (Hepburn et al., 2019; Aresta et al., 2013). Algae cultivation is currently in the early development to early demonstration stages (TRLs 4 to 7). The main benefits of algae cultivation are that it has a high potential for large-scale reuse of CO_2, algal oil can be injected into existing crude oil refineries and the use of algae-derived energy carriers such as biofuel and biogas can displace fossil equivalents. Major barriers of algae cultivation include limitation of capital intensity of the cultivation sys-

Table 11
Estimated market and technology readiness level (TRL) of CO_2 utilisation (Hepburn et al., 2019; Aresta et al., 2013; Patricio et al., 2017; Naims, 2016).

Pathways	Application/ Product	CO_2 Utilised (Mt CO_2/year)	Product Produced (Mt/year)	CO_2 Storage Period	TRL
Direct usage	Algae cultivation for biodiesel	2.0	1.0	Weeks/Months	4 to 7
	Beverage carbonation	2.9	2.9	Days/Months	9
	Enhanced oil and gas recovery (EOR/EGR)	25.0	7% to 23% of oil reserve; <5% of gas reserve	Millennia	9
	Food packaging	8.2	8.2	Days/Months	9
	Industrial gas	6.3	6.3	Days/Months	9
	Urea yield boosting	132.0	180.0	Days/Months	9
Conversion	Carbonates	0.5	>2.0	Decades/Centuries	7 to 8
	Methanol	10.0	60.0	Weeks/Months	7 to 8
	Chemicals (such as formaldehyde and acrylates)	6.5	28.0	Days/Decades	6 to 8
	Polymers (such as polycarbonates and polyurethanes)	1.5	15.0	Months/Decades	7

tems and requirement for large amount of nutrients similar to existing agricultural systems (Global CCS Institute, 2011).

Crop cultivation in greenhouses can be enhance by stimulating the plant growth using high purity CO_2 with some heat (IEA, 2019). For example, the yield of red leaf lettuce can be increased up to 30% by enriching the CO_2 concentration from 200 ppm to 1,000 ppm in the greenhouse (Becker and Kläring, 2016). Flue gases containing impurities such as SO_x, NO_x or heavy metals should not be used in greenhouses as they are toxic to the plants and workers. A suitable CO_2 source for greenhouses is biogas upgrading plants because its CO_2 is sufficient to supply a large greenhouse and it can produce highly concentrated CO_2 streams with excess heat for direct utilisation in the greenhouse. Generally, about 0.50 kg CO_2/h/100 m^2 to 0.60 kg CO_2/h/100 m^2 is required for crop cultivation. CO_2 utilisation in greenhouses is a common practice to increase crop yield and it is a mature technology (TRL 9) (Patricio et al., 2017).

3.4.2. CO_2 utilisation by conversion into products

Captured CO_2 can be used as a feedstock to produce a variety of fuels, chemicals and building materials. Commercially established products such as methane, methanol and syngas can be used directly as a fuel or as an intermediate to produce other fuels such as diesel, gasoline and aviation fuels that are compatible with existing infrastructure. CO_2-derived fuels are mostly applied to the transport sector, for examples, methane can be used for heating and power generation, and methanol can be used as a blend with gasoline if it meets the fuel quality standards. One of the most technologically mature conversion routes is the direct conversion of CO_2 into methane and methanol by methanation and hydrogenation processes, respectively (IEA, 2019).

CO_2 methanation involves the reaction between CO_2 and H_2 to produce CH_4 and H_2O (Equation 12). CO_2 methanation process has high equilibrium conversion between 25 °C and 400 °C and it can reach 99% CH_4 selectivity using suitable catalysts (Li et al., 2018). Park *et al.* (2015) reported that the maximum CH_4 yield from photoreduction of CO_2 with H_2O using TiO_2/5% mol Cu–TiO_2 doubled layered film was twice (175 μmol/g catalyst/L) as that obtained using TiO_2/TiO_2 double layered film (80 μmol/g catalyst/L) (Park et al., 2015). Although methane production process is more energy-intensive than methanol production process, majority of the global CCUS projects still focussed on producing CO_2-derived methane, with almost 70 demonstration plants producing CO_2-derived methane in Germany and other European countries (IEA, 2019).

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \ \Delta H^0_{298} = -165 \text{ kJ/mol} \quad (12)$$

Methanol (CH_3OH) can be synthesised by hydrogenation of concentrated CO_2 over a metal/metal oxide catalyst (such as copper (Cu), zinc oxide (ZnO) and Al_2O_3) at moderate temperature and pressure (about 225 °C at 50 bar) (Equation 13). The H_2 is produced by electrolysis of H_2O, thus making the methanol synthesis process highly energy-intensive. The methanol production process could be economically and environmentally viable if H_2 is produced using electricity from renewable sources that requires either compression at 350 bar to 700 bar or liquefaction at a very low temperature of –253 °C (Patricio et al., 2017). Pérez-Fortes *et al.* (2016) evaluated that a methanol CCU plant utilising 1.37 t CO_2/t methanol would produce 440 kt methanol/year with 94% CO_2 conversion in the process (Pérez-Fortes et al., 2016). They reported that the methanol CCU plant has an electrical energy consumption of 0.61 GJ/t methanol and the plant would be economically viable when the cost of methanol was increased in a factor of almost 2 or the cost of H_2 was decreased by almost 2.5 times or the cost of CO_2 was around USD 255/t CO_2 consumed. Their study showed that such CCU plants have small potential for CO_2 emissions reduction with an estimated net CO_2 emissions reduction of 2.71 Mt CO_2/year if constructed in Europe. Globally, the estimated market for methanol in 2016 was 60 Mt/year with CO_2 utilisation of 10 Mt CO_2/year (Aresta et al., 2013). The first commercial demonstration of CO_2-derived methanol production plant was constructed in Iceland, producing 4 kt methanol annually (Koytsoumpa et al., 2018). Currently, CO_2-derived fuels could not compete with fossil fuels in most regions of the world, nor with many other alternative energy carriers such as direct use of electricity and hydrogen due to low energy conversion efficiency and high capital cost (IEA, 2019; Global CCS Institute, 2011).

$$CO_2 + 3H_2 \rightleftharpoons CH_3OH + H_2O \ \Delta H^0_{298} = -49 \text{ kJ/mol} \quad (13)$$

A wide range of chemicals and materials can be derived from CO_2, for examples, solvents, plastics, fibres and synthetic rubber. Methanol can be converted to olefins (such as ethylene and propylene) for use in polymers production to make plastics and aromatics (such as benzene, toluene and xylene) for use in health and hygiene, food production and processing, information and technology and etc. to make more complex high value chemicals. Currently, the methanol-to-olefins technology is deployed at commercial scale in China and the methanol-to-aromatics technology is still in the demonstration stage (IEA, 2019). A new and low-energy method to manufacture polymers and chemicals was developed by Novomer Inc. in the US, which involves reacting concentrated CO_2 with epoxide (such as ethylene oxide) at low temperature and pressure to produce polycarbonates using a proprietary zinc-based catalyst system. The synthesised polymers are estimated to contain up

to 50% CO_2 by mass. Aliphatic polycarbonates (compounds with carbon atoms linked in open chains) can degrade in 6 months in ideal compost conditions and release CO_2 back to into the atmosphere. CO_2 can be sourced from a waste stream, for examples, flue gas from coal-fired power plants, ethanol fermentation and natural gas wells. Polymers derived in part from CO_2 (such as polycarbonates) could substitute traditional petroleum-based plastics such as polyethylene, polypropylene, polystyrene and PVC and they can be used as surfactants for EOR, coatings for wood and metal in domestic, industry, automotive and food products, and in food and general packaging (Global CCS Institute, 2011). The estimated global market for CO_2-derived chemicals in 2016 was 28 Mt/year with CO_2 utilisation of 6.5 Mt CO_2/year while that for CO_2-derived polymers was 15 Mt/year with CO_2 utilisation of 1.5 Mt CO_2/year (Aresta et al., 2013).

Furthermore, CO_2 can be used as a component of the aggregate (a filler material), as a feedstock in cement (a binding material) production and as an input for concrete curing (IEA, 2019). In carbonate mineralisation (or carbonation) process, moderately concentrated CO_2 (for example, flue gas from power station) reacts with alkaline brine loaded with minerals such as CaO (burnt lime) or MgO (magnesia) to form mineral carbonates such as $CaCO_3$ or magnesium carbonate ($MgCO_3$). Aggregates and supplementary cementitious material are produced and they can be used to make concrete, asphalt and other building materials since carbonates are stable compounds without any leakage concerns. Hence, CO_2 can be stored in the carbonates for a long time, from decades to centuries. The production of 1 tonne of mineral carbonate typically requires 0.5 tonne of CO_2 (Global CCS Institute, 2011). Currently, carbonate mineralisation technology is in its early development stage (TRLs 7 to 8) with about 0.5 Mt CO_2/year being utilised globally to produce >2 Mt/year of carbonates (Aresta et al., 2013). The main advantages of carbonate mineralisation process are that there is no requirement for CO_2 separation or compression, it can be retrofitted to stationary emitters, it is a scalable process and it can capture/remove particulate matter, SO_2, mercury and other metals. However, the carbonate mineralisation technology is likely to be rejected by the cement industry because they manufacture similar products (Global CCS Institute, 2011).

The use of CO_2 in concrete curing (also known as carbonation curing) is an alternative technology to the traditional energy-intensive steam and autoclave curing processes (Patricio et al., 2017). Carbonation curing involves injecting dilute CO_2 from onsite/nearby flue gas sources into the concrete mixture that contains cement, aggregates and H_2O so that the heat and steam requirements in the curing process can be reduced in the production of precast concrete products. This technology is estimated to utilise <120 kg CO_2 for producing 1 tonne of precast concrete with CO_2 sequestered beyond the infrastructure lifetime (Hepburn et al., 2019; Aresta et al., 2013). Although concrete curing technology is currently in small-scale demonstration (TRLs 7 to 8), it has the potential to be commercialised easily due to the existence of concrete curing process in concrete production. Patricio *et al.* (2017) estimated that 96 kt CO_2/year would be consumed using concrete curing technology to produce 800 kt/year of concrete products (Patricio et al., 2017). The main barrier for concrete curing technology is that the concrete sector has limited capital to invest in new technologies as they operate in a highly competitive commodity market (Global CCS Institute, 2011).

3.5. CO_2 storage

The final step in the CCS chain is CO_2 storage. The captured CO_2 are compressed at pressure above 74 bar so that it is either in supercritical or liquified form before it is stored permanently in geological formations (such as depleted or nearly depleted oil and gas reservoirs, saline formations, basalt and ultramafic rocks and deep coal seams) at a depth greater than 800 m and/or deep ocean. Among these CO_2 storage options, storage through CO_2-EOR, in depleted oil and gas fields, and in saline formations are technically mature with TRL between 7 to 9 (Figure 4) (Kearns et al., 2021). Typical geologic characteristics of effective storage sites are rock formations of adequate porosity (millimetre-sized voids/pores) and thickness to provide the capacity for CO_2 storage, sufficient rock permeability to accept the injection rate of CO_2 that will allow CO_2 to move and disperse within the formation, a well-sealed cap rock or barrier at the top of the formation to contain the CO_2 permanently and a stable geological environment to prevent compromising the integrity of the storage site (Global CCS Institute, 2015; Solomon et al., 2008). For a CCS project at US Gulf coast, the indicative costs of CO_2 injection and geological storage in 2020 were ranged from USD 1.72/t CO_2 for onshore good quality geological storage reservoir to USD 18.97/t CO_2 for offshore geological storage reservoir while the indicative costs of CO_2 monitoring and verification in the same year was ranged from USD 1.72/t CO_2 to USD 4.14/t CO_2 (Kearns et al., 2021).

3.5.1. Storage through CO_2-EOR

CO_2-EOR is a mature technology (TRL 9) and it has been widely used in the oil and gas industry for over 50 years (Kearns et al., 2021). In CO_2-EOR process, supercritical CO_2 is injected into nearly depleted oil reservoirs to mix with the oil and reduces its viscosity so that the oil would flow more easily through the rock pore space to producing wells, enabling greater extraction of the oil (Global Energy Institute, 2012). It was reported that EOR (a tertiary recovery process) can extract 30% to 60% more crude oil from the well than the conventional recovery process, which extract 20% to 40% of the crude oil (Cuéllar-Franca and Azapagic, 2015). Although the extraction yield was increased, most dissolved CO_2 returns back to the surface with the pumped oil and some CO_2 may be released into the atmosphere. To reduce the CO_2 purchasing cost, CO_2 is separated from the oil at the surface and recaptured for re-injection so that almost all the CO_2 used will eventually remain in the reservoir permanently at the end of the oil field's life (Hill et al., 2013). This incidental storage of CO_2 allows large quantities of CO_2 to be stored underground safely, securely and for a long period of time (in millennia) (Global CCS Institute, 2015).

Mac Dowell *et al.* (2017) estimated about 70 Gt CO_2 to 140 Gt CO_2 could be injected and stored in more than 90% of the oil reservoirs globally to produce 470 billion barrels (bbl) of additional crude oil (Mac Dowell et al., 2017). The CO_2 utilisation rate for EOR in 2050 was estimated around 0.1 Gt CO_2/year to 1.8 Gt CO_2/year (Hepburn et al., 2019). Rubin *et al.* (2015) reported that power plants with capture, transport and geological storage with EOR credits (i.e., USD 15/t CO_2 to USD 40/t CO_2 sold for EOR) can significantly reduce the overall plant cost and the added cost of CCS by USD 25/MWh to USD 40/MWh for coal-fired power plants and by USD 10/MWh to USD 15/MWh for NGCC power plants when compared with those without EOR credits (Rubin et al., 2015). Based on the CO_2 storage costs in Europe and the US summarised in Table 12, EOR is the most cost-effective CO_2 storage option among the available storage options because it has the lowest costs for CO_2 storage with mean values of USD 0/t CO_2 for onshore storage and USD 6.76/t CO_2 for offshore storage in Europe, and EOR credit of USD 11.44/t CO_2 for onshore storage in the US (Hendriks et al., 2004; Bock et al., 2003). The indicative potential benefit from EOR to CO_2 storage is typically below USD 16/t CO_2 and it can increase up to USD 30/t CO_2 at USD 50/bbl of oil (Metz et al., 2005). EOR would be economically viable if the cost of supplied CO_2 is between USD 45/t CO_2 and USD 60/t CO_2 with oil price at about USD 100/bbl (Godec, 2011; IEA, 2015).

3.5.2. Storage in depleted oil and gas fields

Captured CO_2 can be stored permanently and cost-effectively in depleted oil and gas fields that are no longer economic for oil and gas production (Global CCS Institute, 2015). These reservoirs have established trapping and storage characteristics with a large number of existing equipment installed on the surface and underground that can be reused for CO_2 storage with minor modification (Cao et al., 2020). However, the buoyant plume of injected CO_2 would migrate upwards and

Table 12
CO_2 storage costs for different storage options (Hendriks et al., 2004, Bock et al., 2003).

Storage options	Onshore/Offshore	CO_2 storage cost in Europe (USD/t CO_2 stored)		CO_2 storage cost in the US[a] (USD/t CO_2 stored)
		Range	Mean	
EOR	Onshore	–13.51 to 13.51	0	–11.44[b]
	Offshore	–13.51 to 27.03	6.76	–
Depleted oil field	Onshore	1.49 to 4.86[c]	3.18	3.81[d]
	Offshore	4.86 to 10.41[c]	7.64	–
Depleted gas field	Onshore	1.49 to 4.86[c]	3.18	4.86[e]
	Offshore	4.86 to 10.41[c]	7.64	–
Saline formations	Onshore	2.43 to 7.97[c]	5.20	2.97[b]
	Offshore	6.08 to 15.41[c]	10.75	–
ECBM	Onshore	0 to 40.54	20.27	–5.45[f]

Notes:
[a] Negative cost indicates the cost reduction/credit to the storage system.
[b] Storage depth of 1.2 km.
[c] Storage depth between 1 km and 3 km.
[d] Storage depth of 1.5 km.
[e] Storage depth of 1.6 km.
[f] Storage depth of 0.6 km.

may not be evenly distributed in heterogeneous oil reservoirs if the injected CO_2 is immiscible with oil. In gas reservoir, the injected CO_2 forms a single miscible fluid phase with the natural gas and it is more viscous than natural gas (Solomon et al., 2008). The main issue of CO_2 storage in depleted gas reservoirs is the low reservoir pressure after production (sometimes below 20 bar, a large pressure difference between the reservoir and the transport pipeline at the surface) that may cause a large drop in the reservoir temperature, freezing the residual water, formation of hydrates, fracturing due to thermal stresses and affecting the well injectivity, particularly when cold CO_2 is injected (Twerda et al., 2018, Oldenburg, 2007).

Global storage capacity of depleted oil and gas reservoirs was estimated between 675 Gt CO_2 and 900 Gt CO_2. The storage costs are site-specific and it would increase with the storage capacity if the area has multiple storage sites. The estimated storage costs in Europe were ranged between USD 1.49/t CO_2 stored and USD 4.86/t CO_2 stored for onshore depleted oil and gas fields at depths of 1 km to 3 km and between USD 4.86/t CO_2 stored and USD 10.41/t CO_2 stored for offshore oil and gas fields at the same depths (Table 12) (Hendriks et al., 2004). Meanwhile, the estimated storage costs in the Permian Basin in west Texas in the US were USD 3.81/t CO_2 stored at a depth of 1.6 km for depleted oil fields and USD 4.86/t CO_2 stored at the same depth for depleted gas fields (Bock et al., 2003). These costs could possibly be reduced by reusing existing wells.

3.5.3. Storage in saline formations

Another technically mature CO_2 storage technology is storage in saline formations (TRL 9) due to its huge storage capacity (between 1,000 Gt CO_2 and 10,000 Gt CO_2) and wide availability (onshore and offshore, usually located near to CO_2 emission sources) over other storage options. Saline formations are deep underground porous reservoir rocks saturated with brackish water or brine, typically 700 m to 1,000 m below ground level (Metz et al., 2005). CO_2 can be stored in saline formations by hydrodynamic, residual, solubility and mineral trapping mechanisms. Hydrodynamic trapping occurs when CO_2 is trapped as supercritical fluid/gas under a low permeability caprock. However, this method of CO_2 storage is highly dependent on the sealing capacity of the caprock, which is a challenge when selecting the storage site. Residual/capillary trapping occurs when large amount of immobilised CO_2 is trapped in small clusters of pores, which can limit significantly the movement of injected CO_2. More CO_2 can be trapped as residual gas if the sweeping efficiency is increased by increasing the ratio of viscous to gravity force and the heterogeneity in saline formations.

Solubility trapping occurs when CO_2 dissolved in the formation fluid until it reached equilibrium, which may take thousands of years for CO_2 to be completely dissolved in brine due to very small molecular diffusion coefficient. A possible approach to increase the storage capacity in reservoir is dissolution-diffusion-convection process, which requires more study to verified its capability. Mineral trapping occurs when CO_2 reacted with minerals present in the formation to form stable, solid compounds such as carbonates. Mineral trapping is only significant at geological time scale because mineral dissolution is a very slow process (Zhang and Song, 2014). Solubility and mineral trapping are important for saline formations without lateral seals. Generally, the main issue of storing CO_2 in saline formations is the pressure build-up and CO_2 plume migration in saline formations that can fracture the reservoir rock and may damage the top seal as well as reactivate pre-existing fractures and faults through which CO_2 can leak (Orlic, 2016).

The estimated CO_2 storage cost for onshore saline formations in Europe for depths of 1 km to 3 km were between USD 2.43/t CO_2 stored and USD 7.97/t CO_2 stored, which was lower than that for offshore saline formations for the same depth (i.e., between USD 6.08/t CO_2 stored and USD 15.41/t CO_2 stored) (Table 12) (Hendriks et al., 2004). Bock *et al.* (2003) estimated that the storage cost for onshore saline formations in the US was USD 2.97/t CO_2 stored at a depth of 1.2 km (Bock et al., 2003). These costs showed CO_2 storage in saline formations in Europe was more expensive than in depleted oil and gas fields in the same region for both onshore and offshore storage. It also showed CO_2 storage in onshore saline formations in the US was comparable to depleted oil and gas fields in the same area.

3.5.4. Storage in basalt and ultramafic rocks

Unconventional formations such as basalts and ultramafic rocks have CO_2 storage potential through carbon mineralisation. Around 90% of injected CO_2 can be mineralised in these rock formations as a stable carbonate in a few months to decades and they can store up to 60,000,000 Gt CO_2 globally since there are huge area of basaltic rock found in ocean basins worldwide and some onshore areas such as in Washington State in the US, Russia and India, and ultramafic rocks found in Iceland and southern Washington in the US (Kelemen et al., 2019; National Petroleum Council, 2019b). CO_2 is usually injected into hydrologically fractured basalt or permeable zones between basalt flows as basaltic rock has very low permeability.

The main issues with this type of CO_2 storage are potential formation of a silica passivation layer that may decrease the rock reactivity over time and filling of pore space with reaction products that may decrease the porosity, which will lead to lower permeability and reduced fluid

supply (Romanov et al., 2014; Xing et al., 2018). Currently, there are 2 pilot projects (i.e., Wallula Project in the US and CarbFix project in Iceland) that injected and stored CO_2 into basalts for mineralisation. However, the injection rates into these rock formations are still low and most of the tools for conventional CCS cannot be applied to monitor CO_2 plume in these rock formations. Due to these reasons, this storage technology is still in the research stage (TRL 3) (Kearns et al., 2021).

3.5.5. Storage in coal seams through CO_2-ECBM

Another unconventional CO_2 storage option is in coal seams through ECBM. This technology is still in the research stage (TRL 3) because more further studies and trails are required to confirm its storage feasibility. CO_2-ECBM involves flooding partially depleted coal seams with injected CO_2, where it is adsorbed on the porous coal, to displace CH_4 (natural gas), which is released to the surface for it to be captured and consumed as fuel. CO_2 utilisation rates depend on the nature of the coal seam, storage ratio (i.e., the ratio of CO_2 adsorbed to coal bed methane desorbed) and pressure of the coal seam. The storage ratio is about 2:1 for high volatile bituminous coals at low to medium pressure and about 8:1 for lower quality coals at the same pressure. For lignite (a low-quality coal), has a storage ratio of 13:1 (Global CCS Institute, 2011). Bachu (2007) estimated about 800 Mt CO_2 could be stored in the coal beds in Alberta, Canada for an economic CO_2 storage capacity threshold of 200 kt CO_2/km^2 (Bachu, 2007). The storage capacity of unmineable coal formations is uncertain and it was estimated between 3 Gt CO_2 and 200 Gt CO_2 globally with most coal bed methane-producing wells less than 1 km depth. CO_2-ECBM can increase CH_4 production by 90% while conventional recovery from coal beds is only 50% (Metz et al., 2005).

The main issue with ECBM is the reduced permeability of the coal during CO_2 injection due to plasterisation and swelling of the coal that will reduce the size and connectivity of the fractures, hence additional wells are required and this will increase the costs and operational complexity (Kearns et al., 2021). Currently, CO_2-ECBM is still not yet commercially feasible. Bock *et al.* (2003) estimated that onshore CO_2-ECBM storage of 2.2 Mt CO_2/year in a coal bed in the US at 0.6 km depth would require 270 wells with a net storage cost of –USD 5.45/t CO_2 stored (Bock et al., 2003). This CO_2-ECBM storage costs in the US was comparable to the CO_2-ECBM in Europe, which was estimated between USD 0/t CO_2 stored and USD 40.54/t CO_2 (Hendriks et al., 2004). When compared to CO_2-EOR, the storage cost for CO_2-ECBM is not cost competitive in both Europe and the US (refer to Table 12).

3.5.6. Storage in deep ocean

Oceans are the biggest natural CO_2 sink as it covers over 70% of the earth's surface and estimated to have a storage capacity of 38,000 Gt CO_2 with an annual CO_2 uptake rate of about 1.7 Gt from the atmosphere (Leung et al., 2014). Ocean storage of CO_2 may cause ocean acidification that may affect the marine ecosystem. Barry *et al.* (2004) reported high rates of mortality for infaunal deep sea organisms (such as flagellates, amoebae, and nematodes) that inhibit sediments near to the sites of CO_2 release from the seafloor at 3.6 km depth in California in the US due to large changes in seawater chemistry with pH reductions of about 0.5 to 1 (Barry et al., 2004). Moreover, Rae *et al.* (2018) found rapid (in millennial to centennial scale) drop in pH during sudden increase in atmospheric CO_2, indicating rapid transfer of carbon from the deep ocean to the upper ocean and the atmosphere (Rae et al., 2018). A possible approach could be water column injection technology that involves the injection of a very dilute initial CO_2 into the deep ocean through a series of diffusers to minimize local changes in seawater chemistry (Metz et al., 2005).

Generally, the captured CO_2 can be injected into the deep ocean through a pipeline laid on the seabed or through tanker. The first option involves transporting pressurized liquid CO_2 through a pipeline laid on the seafloor and injecting it to a depth where it will be effectively stored for centuries to millennia. If CO_2 is injected through a diffuser at intermediate depths of 1 km to 2 km, the liquid CO_2 droplet plume will rise by buoyancy while dissolving in seawater and it will flash into vapour and bubble up to the surface before it reaches a depth of 0.5 km. If CO_2 is injected at depths greater than 3 km, the liquid CO_2 will be denser than seawater and it will sink to the ocean bottom. Although the technology is available for ocean storage through pipeline, its technical and economic feasibilities, and environmental consequences on the ocean are not well studied and understood. The second option involves transporting CO_2 by refrigerated tanker from a port facility to an offshore floating platform, where CO_2 is injected into the deep ocean through a vertical pipe. So far, ocean storage through tanker technology is not commercial applicable. The estimated levelized annual cost of CO_2 storage in the deep ocean through subsea pipeline from shoreline to a depth of 2 km, at which CO_2 is discharged via a diffuser unit was between USD 2.90/t CO_2 avoided and USD 14.23/t CO_2 avoided, including transportation, injection and monitoring costs. Meanwhile, the estimated levelized annual cost of CO_2 storage in the deep ocean by three 22,000 m^3 tankers that supply 22 kt CO_2/day to the shoreline collection point with a vertical pipe for CO_2 injection to a depth of 2 km was between USD 15.76/t CO_2 avoided and USD 22.79/t CO_2 avoided, including transaction, transportation, injection and monitoring costs (Heddle et al., 2003).

4. Conclusions

This article has provided a technical and economic review on CCUS systems to assess their current development progress in achieving a net-zero CO_2 emissions future. The opportunities and challenges in various technologies associated with CO_2 capture, separation, transport, utilisation and storage have been discussed. Many of the technologies are considered potential for reducing CO_2 emissions but the global deployment of CCS/CCUS projects is still not fast enough to meet the net-zero CO_2 emissions target by 2050. A possible solution for this could be the development of a hybrid CCS/CCUS system involving two or more processes for CO_2 capture, separation, transport, utilisation and/or storage. This novel approach can offer combined technical benefits from the processes, which can possibly improve the overall system performance, particularly in terms of energy efficiency and cost. However, this approach needs to be further investigated to evaluate and quantify their technical and economic viability for industrial applications.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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