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Critical Analysis and Evaluation of the Technology Pathways for Carbon Capture and Utilization

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Abstract: Carbon capture and utilization (CCU) is the process of capturing unwanted carbon dioxide (CO_2) and utilizing for further use. CCU offers significant potential as part of a sustainable circular economy solution to help mitigate the impact of climate change resulting from the burning of hydrocarbons and alongside adoption of other renewable energy technologies. However, implementation of CCU technologies faces a number of challenges, including identifying optimal pathways, technology maturity, economic viability, environmental considerations as well as regulatory and public perception issues. Consequently, this research study provides a critical analysis and evaluation of the technology pathways for CCU in order to explore the potential from a circular economy perspective of this emerging area of clean technology. This includes a bibliographic study on CCU, evaluation of carbon utilization processes, trend estimation of CO_2 usage as well as evaluation of methane and methanol production. A value chain analysis is provided to support the development of CCU technologies. The research study aims to inform policy-makers engaged in developing strategies to mitigate climate change through reduced carbon dioxide emission levels and improve our understanding of the circular economy considerations of CCU in regard to production of alternative products. The study will also be of use to researchers concerned with pursuing empirical investigations of this important area of sustainability.

Keywords: carbon capture and utilization (CCU); carbon dioxide; technology pathways; critical analysis and evaluation; circular economy

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1. Introduction

Carbon capture and utilization (CCU) refers to the process of capturing unwanted carbon dioxide (CO_2) in order for it to be recycled for further use [1,2]. This is distinct from carbon capture and storage (CCS), where the CO_2 is captured and compressed as pressurized gas for long-term storage at geological sites [3]. Both CCS and CCU offer potential technological solutions to mitigate the deleterious impact of climate change caused by excessive burning of hydrocarbons and the resultant greenhouse gas (GHG) effect of high levels of carbon dioxide (CO_2). In this context, it has been identified that urgent action is needed in order to ensure that global warming does not exceed 1.5 °C above pre-industrial levels [4] and this is an extension of the earlier obligations established in the Paris Climate Conference of 2015, where 195 countries agreed to implement a legally binding agreement and corresponding action plans to work towards limiting global warming to below 2 °C [5]. In the case of limiting the increase to 2 °C, it has been estimated that global CO_2 emissions would need to be reduced by approximately 50% by the year 2050 [6] and hence there is now an urgent need to implement solutions that have the capacity to be ramped up rapidly to industrial scale in order to achieve this magnitude of reduction in CO_2 levels. It should be noted, however, that the original 2 °C target is subject to a range of assumptions and probabilities associated with the possible reduction in CO_2 levels. For instance, pursuing a course of action through limiting global warming caused by anthropogenic CO_2 emissions alone, according to a probability of >33%, >50%, and >66% to less than 2 °C since the period 1861–1880, will potentially need the cumulative CO_2 emissions that arise from all of the anthropogenic sources to stay between the values of 0 and about 1570 GtC (5760 $GtCO_2$), 0 and about 1210 GtC (4440 $GtCO_2$), and 0 and about 1000 GtC (3670 $GtCO_2$), respectively, and since that period [6]. The original target can be viewed in terms of a complex system related to many inter-related factors and also being subject to achievement of political consensus, which builds on the underlying scientific assessment [7]. However, clearly staying within the increase of up to 2 °C is an important target in the context of mitigating the impact of climate change although it is recognized that staying within an increase of up to 1.5 °C is the preferred outcome, which is clearly a more demanding target to be pursued.

Indeed, both CCU and CCS offer potential benefits in regard to responding to the impending threat of climate change through reducing the level of carbon dioxide emissions [8]. This will help to underpin achievement of the aforementioned international targets alongside adoption of other renewable technologies, such as solar, wind, tidal and hydroelectric power generation [9] as well as the transition to electric vehicles for transportation applications [10]. Despite the fact that CCU is likely to have a potentially lower capacity level than CCS for reducing carbon dioxide (CO_2) levels, it does nevertheless offer a route to reduce CO_2 emissions as well as providing a process to develop value added materials for further use as part of the circular economy paradigm. Although it should be noted that where CO_2 is subsequently incorporated into materials that gradually oxidize over extended periods, the resulting CO_2 emissions still take place. In such a case, the CCU process cannot be regarded as carbon neutral and is more accurately described as a temporary storage of carbon that still results in medium-to-long term emissions. It is worth also noting that a complementary approach to CCU is direct air capture of carbon dioxide and various researchers have examined different aspects of this process, including the suitability of materials able to adsorb CO_2 directly from air and other gas mixtures [11] as well as reduction in the cost of calcium-based direct air capture of CO_2 [12].

It has been calculated that adoption of CCU technologies has the potential to lead to reductions in reduce GHG emissions by up to 3.5 Gt CO_2-equivalent in 2030 [13] through effectively decoupling chemical production from fossil resources, although this would be dependent on addressing the need for largely increased mass flows and the resultant demand for low-carbon electricity. It should be added that this estimate was generated through a modelling process based on scenario analysis, where different scenarios were considered. These scenarios involved effectively decoupling chemical production from fossil resources through using captured carbon dioxide as the feedstock for production of various chemicals, fine chemicals and polymers. This would represent a highly disruptive course of action to be followed by the chemical industry that would require major levels of capital investment and consequently the scenario analysis needs to be treated with an appropriate level of caution. Nevertheless, it does highlight the potential scope for CCU to be deployed for this application, albeit due to certain assumptions and supporting scenarios being followed. A further illustration is that it has been estimated (as of 2011) that through replacing around 10% of building materials with carbonate minerals would have led to an expected reduction in CO_2 emissions by 1.6 Gt/year, which at the time was about 5% of the global CO_2 emissions [14]. CCU also provides a possible pathway towards a circular economy [15] through recycling waste CO_2 and the resultant conversion into higher value materials, such as concrete [16], organic compounds [17] and polymeric materials [18].

The structure of this article is as follows. After the introduction is the literature review, which establishes the background and industrial requirements for CCU. This is followed by the method section and thereafter the section that details the analysis and evaluation activities that have been undertaken. The next section provides the value chain analysis, followed by conclusions and future work.

2. Literature Review

There are a range of technology applications where CCU has been investigated by researchers. These include diverse areas, such as CCU based on chemical conversion of CO_2 from industrial flue gas under ambient conditions [19], CCU via CO_2 conversion to liquid transportation fuels through reacting with renewable hydrogen produced from solar water splitting [20], CCU enabled via a sorptive reactor for CO_2 capture and subsequent conversion to renewable methane [21], CCU as a potential route to plastic products for consumer applications [22], CCU as part of the mineral carbonation process [23], CCU potential in the steel industry [24], CCU for sodium bicarbonate production via solar thermal power [25], wastewater treatment processes as a source for CCU [26], CCU through the fermentation of CO_2 via integration of ethanol fermentation with succinic acid production [27], and CCU in a microalga-based bio-refinery [28]. This range of areas highlights the sheer breadth of research avenues that are currently being explored in this area—across power generation applications to the production of novel products and materials as well as different industrial settings and novel bioengineering applications.

In order for CO_2 to be utilized for industrial applications, there is first a need to capture the CO_2 [29]. From the relatively more established perspective of CCS, there have been three main approaches available in regard to capturing CO_2 from power generation plants [30], including pre-combustion capture, post-combustion capture and oxy-fuel capture, as well as the capture of CO_2 from industrial processes. There continues to be investment into the development of carbon capture technologies in support of both CCS and CCU (collectively known as carbon capture, utilization and storage, or CCUS). There are, however, various issues that need to be addressed in regard to key policy determinants as well as the various economic, technological and environmental issues. Specifically in the case of CCS [31], certain decision factors have been identified as having a high impact (namely capture technology, storage technology, and cost reduction), others a moderate impact (namely investment decision, environmental assessment, regulatory framework, and site selection) and some with a low level impact (namely transportation system, public awareness, government funding, monitoring technology, and international collaboration). Although these factors relate principally to CCS and not CCU, they do nevertheless provide an indication of where the priorities lie in terms of further developmental needs for the underlying technologies associated with both processes.

In terms of optimizing carbon capture technologies, researchers have investigated different strategies and approaches, for instance, the process design and economic aspects of capturing carbon dioxide from the flue gas generated by a power plant using experimental data [32]. This study found that deploying pressure swing adsorption can be viewed as a potential carbon capture technology, since increasing the pressure can lead to a higher level of carbon dioxide adsorption. This was undertaken in the context of exploring the utilization of CO_2 in the production of aerogel in order to investigate application as a nano-based thermal insulator. A comparative analysis of the aerogel along with other building insulator materials highlighted that the level of energy saving as well as avoided CO_2 are higher in the case of aerogel. In other work, synthetic hydrocarbons can be produced in a sustainable manner via power-to-gas processes [33], thereby leading to an overall net reduction in greenhouse gas emissions that is a result of substituting conventional natural gas and other hydrocarbons with carbon neutral alternatives. This scenario-driven approach was developed according to a node-based model as part of a case study investigation in Finland. The framework adopted enables assessment of a range of CO_2 utilization implementation strategies through considering different CO_2 storage and transport scenarios. The study found that CO_2 capture costs were significant in some scenarios and in one case the cost of storage was approximately four times more costly than the baseline scenario (i.e., 354 M€ and 85 M€, respectively). This highlights the economic impact of the CO_2 storage and transport arrangements that are implemented for a given CCU facility. Moreover, CO_2 sources with smaller annual emissions appears to increase capture costs by 14% compared to the baseline scenario. This increase in cost can be viewed as being equivalent to the transportation cost for over a quarter of all the CO_2 that is captured through off-site processing (varying distance, 100–400 km). The studies are particularly insightful in regard to developing a matrix to support key decisions for a CCU system and according to the parameters of supply, demand, storage and transport versus scale, type, units, location and technology.

Implementation of CCS and CCU projects will also rest on the adoption of viable options for the transport of CO_2 from large facilities such as power plants and industrial facilities [34]. On this matter, studies have proposed that pipeline transport of liquid CO_2 would be the most economically viable method for transporting large volumes of CO_2 [35]. In this case, it has been calculated for a case study in the Midwest of the United States there would be a pipeline cost of USD $1.16 per tonne of CO_2 transported for a 100 km pipeline that can handle 5 million tonnes of CO_2 per year (this represents an approximate output of an 800 MW coal-fired power plant fitted with carbon capture technology). Although the modelling approach is subject to certain assumptions and sensitivity analysis, it is nevertheless a useful approach to help understand the economic conditions of transporting captured liquid CO_2 as part of assessing the commercial viability of future CCS and CCU schemes and projects.

Adoption of new energy related technologies not only rests on technological, economic and environmental factors but there is also a need to consider the level of public awareness, perception and acceptance of the adoption of new technologies (i.e., social considerations in the context of the triple bottom line of sustainability) [36]. In the case of CCS and CCU, studies conducted in Germany identified that although both technologies were viewed as generally being accepted, nevertheless CCU was viewed significantly more positively than the case of CCS. This social science-based study according to an online survey instrument (N = 509) found that acceptance of CCS was negatively influenced by the risks associated with storage and transport, while for CCU the disposal and product risks appeared to decrease the level of acceptance. This work is important in regard to understanding how to communicate the benefits of new technologies (such as CCS and CCU) in order to address public and social-based concerns, and thereby support policy positions in this area as well as governmental support mechanisms for technology adoption (such as R&D funding and tax credits for appropriate technology investments). Moreover, there is a need to ensure effective knowledge transfer mechanisms are available to support adoption of renewable energy technologies and this has been identified as a key requirement for CCU projects in the Philippines [37].

In regard to adopting an environmental perspective of CCU, life cycle assessment (LCA) can be a useful tool to quantify environmental metrics [38–40], although there is the ability to generate different results according to the boundary conditions and supporting assumptions adopted in the research study [41]. Since CCU can be considered as both an energy intensive as well as material intensive process, it is not entirely evident whether or not the process will eventually result in a net reduction in environmental impacts according to a life cycle perspective. Indeed, researchers have investigated the consequential life cycle assessment of carbon capture and utilization technologies within the German chemical industry implemented for polyurethane and methane production [42]. The analysis was undertaken with the openLCA 1.7.3 and Brightway2 LCA software enabled through access to background data from the ecoinvent database 3.4 consequential. The research revealed that in both the near- and a long-term scenarios, the global warming impact for all CO_2 conversion technologies is negative, apart from the cases of dimethoxymethane, electrochemically produced formic acid, and Fischer–Tropsch production. The researchers also identified the conversion technologies that have the highest potential for enabling a reduction in the global warming impact from a life cycle perspective are formic acid produced via hydrogenation and polyol production. It was concluded by the authors that there is merit from an environmental viewpoint in developing CCU technologies for use in the chemical industry. Other researchers have reported on the methodological challenges with applying LCA to CCU [43]. This includes where utilized CO_2 may be considered as leading to negative GHG emissions due to the CCU process resulting in products in both the capture and utilization stages and also the lack of consideration of the CO_2 storage duration. Therefore, a systematic process for LCA of CCU was developed where utilized CO_2 is considered as regular feedstock with its own corresponding production emissions.

CCU includes a number of technologies that are generally at an early stage and require further development as well as the need to overcome commercial and regulatory barriers [44]. Nevertheless, the utilization of CO_2 does support the achievement of climate change goals (such as the sustainable development goals) if the application can be scaled-up according to the required capacity levels. In some cases, there is also the potential for semi-permanent carbon retention, for instance in the case of the production of building materials. There are presently, however, a number of challenges associated with CCU [45]. These include technological and economic issues as well as the need to understand in more detail the full life-cycle benefits of utilizing CO_2 in different applications from an environmental perspective. Consequently, it is useful to conduct a critical analysis and evaluation of the technology pathways for CCU in order to investigate the potential engineering solutions according to the features and trends of this emerging area of sustainability and clean technology.

3. Method

Figure 1 provides an illustration of the methodological scheme adopted for the research study based on the critical analysis and evaluation of the technology pathways for CCU. Critical analysis and evaluation is a recognized methodology that is employed in order to support assessment of complex situations through considering the extant literature on a subject and augmenting this consideration with further analysis and evaluation of the main factors identified by the research study. This approach is suitable since CCU is still an emerging area of sustainability that is yet to be fully investigated and hence a study that is exploratory in nature is appropriate in this case. Example research studies that have adopted this approach include the measurement of circular economy strategies via index methods that are enabled through via a critical analysis approach [46], critical analysis of green building research trends [47], and the critical analysis of IoT (internet of things) as a system for elderly monitoring [48].

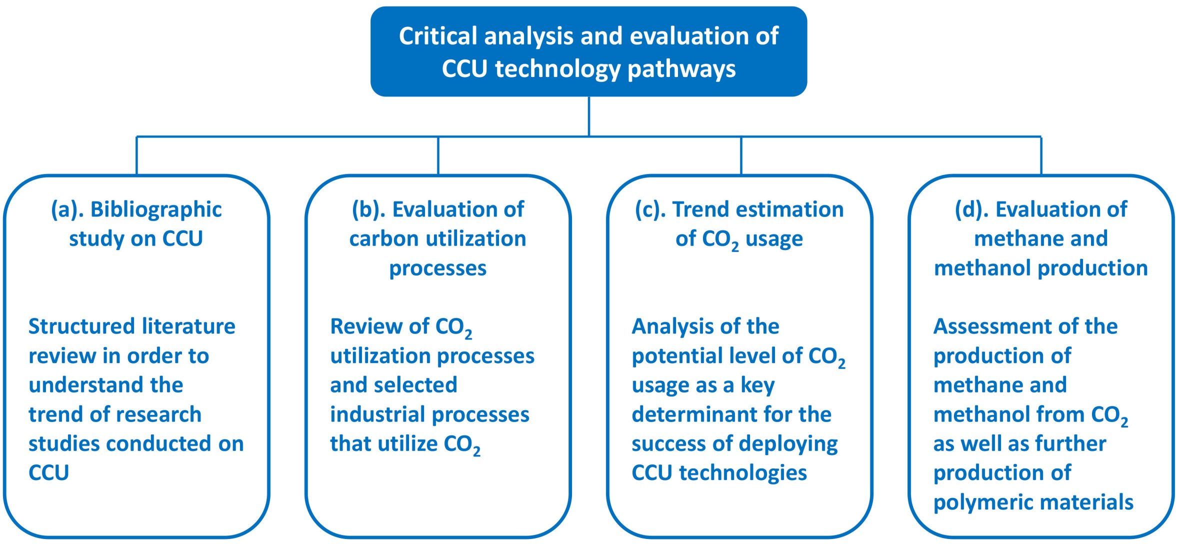

Figure 1. Methodological scheme adopted in the research study.

The scheme involved four main strands of investigation. The first was a bibliographic study of CCU in order to develop an improved understanding of the maturity of the research field and underlying publication trends. The second involved review of key sources of information as part of an evaluation of carbon utilization and corresponding industrial processes. The third involved trend estimation of CO_2 usage, including analysis of global CO_2 demand, global urea production and global EOR (enhanced oil recovery) levels. The fourth includes an assessment of the production of methane and methanol from CO_2 as well as the scope for eventual conversion to polymeric materials, such as polyoxymethylene (POM).

4. Results

4.1. Bibliographic Study on CCU

In order to understand the trend of research studies conducted on CCU, a literature search was conducted on 26th November 2020 using the ScienceDirect electronic database. This database was selected because it contains a large number of publications related to science and engineering areas. The search term used was: carbon capture and utilization, which was searched in the field of title, abstract or author-specified keywords for the period used, which was 2010 to 2019 (10-year period). This resulted in 1292 publications over this period. The number of CCU focused articles according to year of publication is provided in Figure 2. Furthermore, where the field was the entire article, the number of publications was a much higher figure of 79,331 articles, although a large proportion of these results did not focus on CCU in any great detail and hence it is appropriate for the purposes of this study to concentrate on the results of the aforementioned search that resulted in 1292 articles. For comparative purposes, a similar search was performed over the same 10-year period (2010–2019) in the ScienceDirect database in the field of title, abstract or author-specified keywords for the search term: carbon capture and storage. In this case the number of articles was a much higher figure of 3596 (*cf.* 1292 for carbon capture and utilization). This highlights that the research field for the related area of CCS is considerably larger and more mature than the CCU research field. Nevertheless, CCU remains a worthy area of technological development to be investigated according to a systematic and evidence-based approach according to critical analysis and evaluation of the technology pathways for this industrial process.

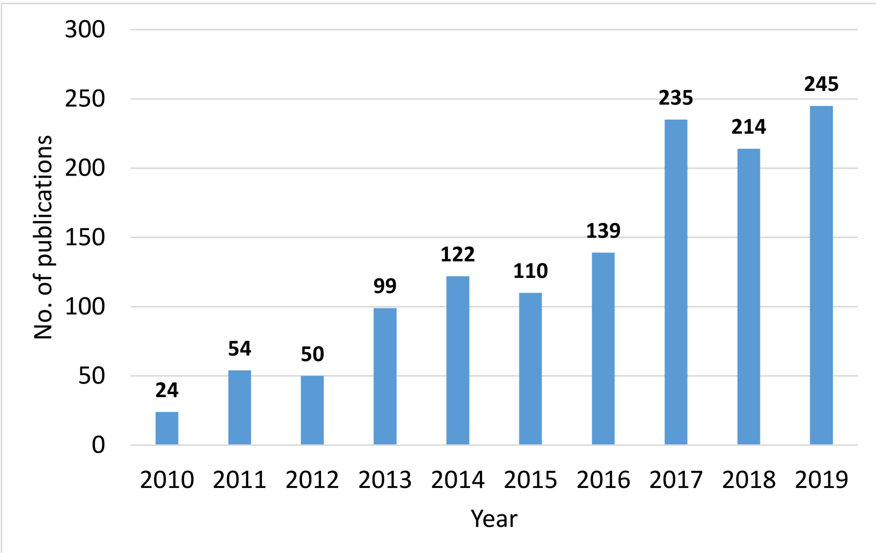

Figure 2. Number of carbon capture and utilization (CCU) focused articles according to year of publication.

Figure 2 shows that over the period of 2010 to 2019, there has been a steady increase in the number of articles focused on CCU, with the highest number appearing in the most recent full year of 2019 (N = 245). In 2010, there were 24 articles and this figure increased steadily over the 10 year period, although there was a small dip from 2011 (N = 54) to 2012 (N = 50) as well as larger jumps from 2012 (N = 50) to 2013 (N = 99) and from 2016 (N = 139) to 2017 (N = 235). This overall trend highlights that CCU is becoming a more popular area of research and as such CCU can also be considered as an emerging area of industrial technology. Figure 3 provides the number of articles for the main types of publications, with the highest number being research articles (N = 1085). This is consistent with CCU being an area of emerging technology that requires a significant amount of primary research to be conducted in order for the technology to be adequately developed, evaluated, validated, tested and ultimately deployed.

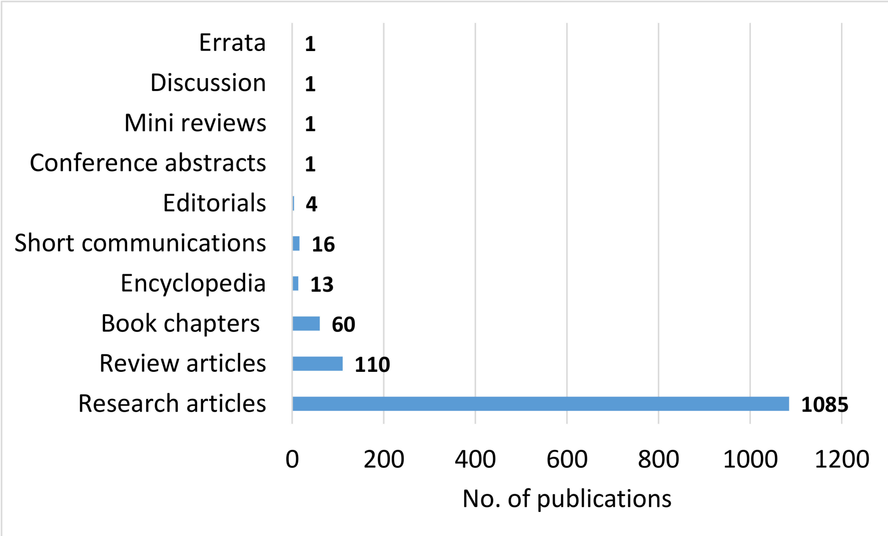

Figure 3. Number of articles for the main types of publications.

Data was captured on the number of articles for the most popular publications (N = 874), with the most frequent publication being Energy Procedia (N = 186), which is an open access collection of conference proceedings spanning the field of energy science, technology and engineering. This data is provided in Figure 4. Other academic journals that featured a high number of articles focused on CCU from the search included the International Journal of Greenhouse Gas Control (N = 107), Applied Energy (N = 85), Energy (N = 57), Journal of Cleaner Production (N = 49) and Journal of CO_2 Utilization (N = 49). This highlights where publications on CCU technologies are mainly appearing.

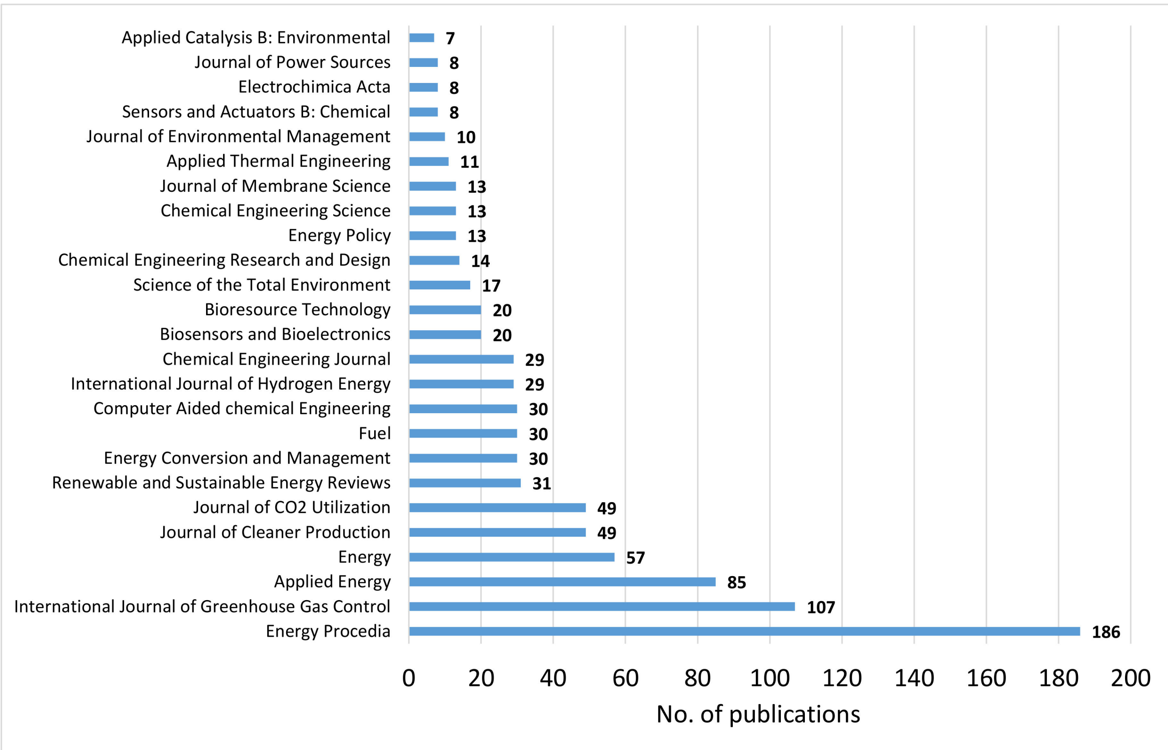

Figure 4. Number of articles for the most popular academic publications.

Finally, the bibliographic search identified the subject areas for the publications. Since articles can be associated with multiple subject areas, this generated an overall number of subject areas of 2415. This data was converted into percentages and is provided in Figure 5, which is a pie chart showing the percentage distribution of the articles focused on CCU according to the main disciplinary or subject areas. This highlights that the highest level of subject areas covered by the articles focused on CCU were, not surprisingly, associated with the area of energy (32%), followed by environmental science (14%), materials science (14%), chemical engineering (13%), engineering (10%), earth and planetary sciences (7%), chemistry (6%), biochemistry, genetics and molecular biology (2%), computer science (2%), and agricultural and biological sciences (1%). This readily identifies the field of research studies on CCU to be highly multidisciplinary in nature.

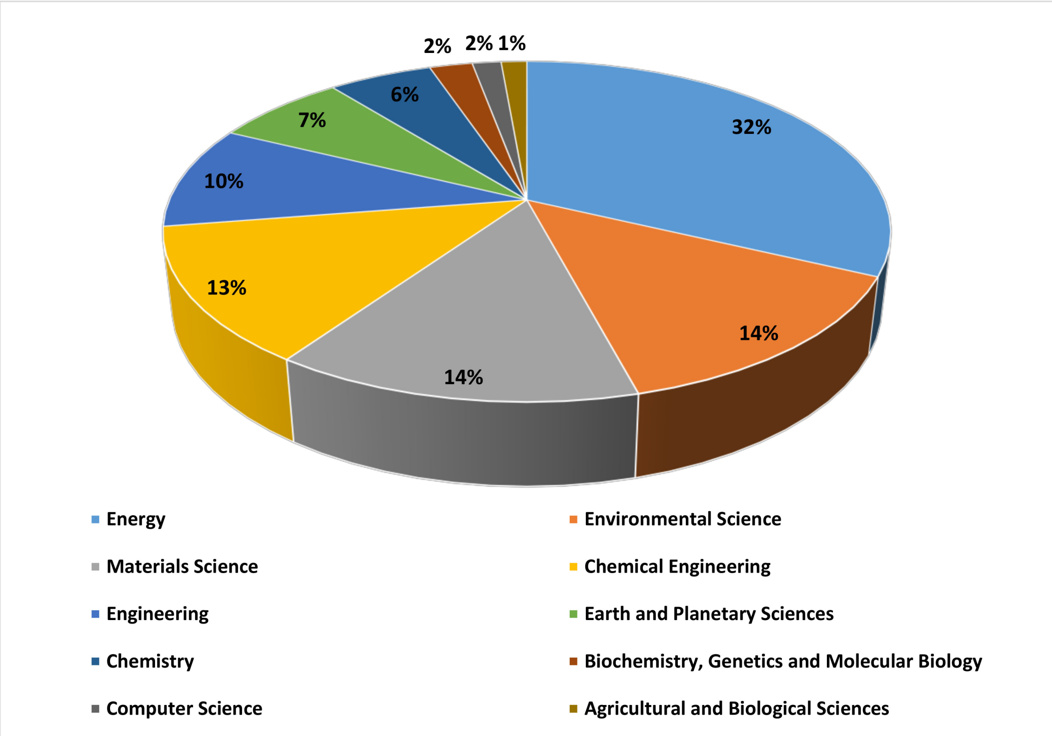

Figure 5. Pie chart showing the percentage distribution of the articles focused on CCU according to subject areas.

4.2. Evaluation of Carbon Utilization Processes

There are a number of potential options for waste carbon dioxide to be utilized, as identified by Al-Mamoori et al. [49]. In this scheme, there are four main processes available, which include enhanced recovery of oil and gas, chemical conversion, mineralization and desalination. Although it is recognized that while desalination has value as a process, it does not lead to the removal of waste carbon dioxide and therefore has not been considered further as part of this current analysis. Enhanced recovery of oil and gas refers to enhanced oil recovery (EOR) and enhanced gas recovery (EGR), which have both been undertaken for many years and have historically represented a major use of CO_2 [50]. As part of the EOR process, CO_2 is injected into the depleted oil well in order to maximize the level of production, where the injected CO_2 mixes with the oil in hard-to-recover rock formations. The mixture of oil and CO_2 is subsequently pumped to the surface, where any excess CO_2 can be recycled back into the cycle and the process repeated, which ultimately leads to more barrels of oil being recovered when compared to traditional methods.

Chemical conversion can involve chemically converting CO_2 into a number of different products and intermediate feedstock materials, including methane and methanol [51], as well as leading to syngas and other alkanes [52]. Two key processes in this context for the production of hydrocarbon fuels are CO_2 hydrogenation [53] and the dry reforming of methane (DRM) [54]; in the latter case, the high operating temperatures of the process result in difficulties in finding an appropriate catalyst for the reaction. Carbon dioxide can also be converted into a range of fine chemicals, including urea, inorganic carbonates, polyurethane, acrylic acid, acrylate polycarbonates and alkylene carbonates. Further CCU processes in this scheme include mineralization into carbonates, which unfortunately has a number of chemical process challenges.

It is useful to build on the scheme by Al-Mamoori et al. [49] by removing the category of desalination (due to the nature of the process not leading to removal or conversion of CO_2) and adding a new category, which is the direct synthesis of CO_2-based polymers (see Figure 6). This category differs from the chemical conversion category since it primarily involves conversion to feedstock materials, which are then further converted to other materials (including polymers). The new category includes a range of synthetic methods and technologies that are currently under development. For instance, synthesis of elastomers through the reaction of carbon dioxide with propylene oxide and maleic anhydride. In this example, the resulting cross-linkable polyether carbonate polyols are combined with isocyanates to lead to CO_2-based rubbers, which can be considered as a novel class of polymers [55]. Other studies have identified CO_2 as a possible feedstock for the production of oxalic and glycolic acid based polyesters [56] as well as the synthesis of CO_2-based polymers, co-polymers and polymer blends, such as CO_2-based aliphatic polycarbonates and poly (propylene carbonates) [57]. These synthetic pathways are still at the exploratory stage of development but nevertheless there are clear opportunities for further studies to evaluate the technological and economic case for production of polymeric materials directly based on captured carbon dioxide.

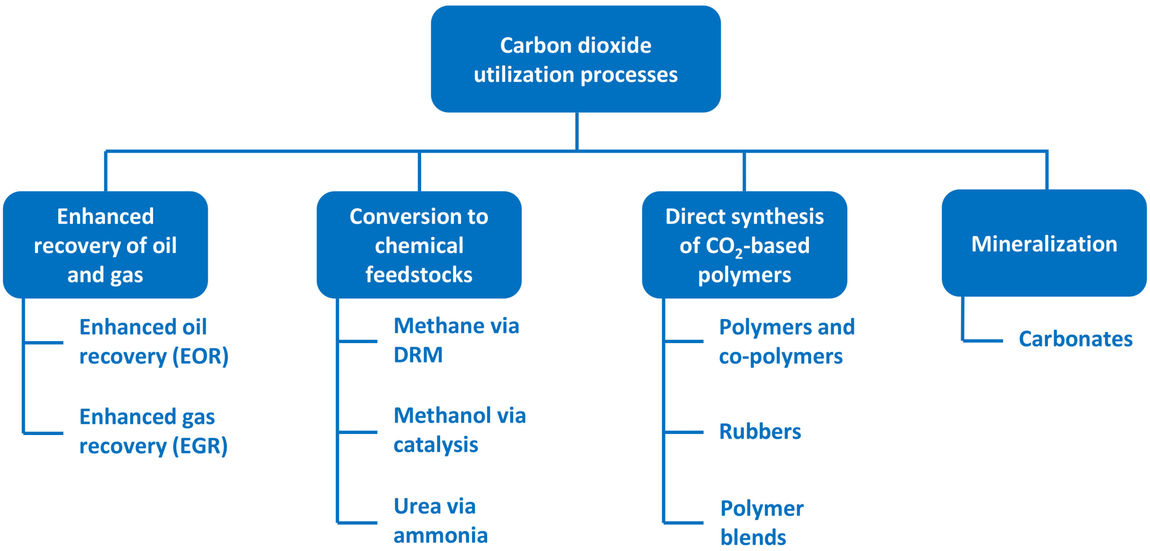

Figure 6. Carbon dioxide utilization processes, adapted from Al-Mamoori et al. [49].

In consideration of the use of CO_2 in industrial processes, there are a range of technologies and corresponding products that are currently under development that have been evaluated by Patricio et al. [58] as follows:

- Bauxite residue carbonation where CO_2 is used to neutralize bauxite residues (TRL = 9).
- Horticulture production based on CO_2 supplementation via plant growth (TRL = 9).
- Urea production from ammonia and CO_2 (TRL = 9).
- Concrete curing of concrete blocks where CO_2 used for precast concrete curing (TRL = 7–8).
- Mineral carbonation based on CO_2 reacted with calcium or magnesium containing minerals (TRL = 7–8).

- Lignin production where CO_2 is used in black liquor pH regulation (TRL = 7–8).
- Methanol production based on electrochemical reduction in CO_2 (TRL = 7).
- Polyurethane production where CO_2 is reacted with calcium or magnesium containing minerals (TRL = 7).
- Polycarbonate production where CO_2 is used as raw material to produce plastics and fibres (TRL = 7).

This evaluation involved an assessment by the researchers of the technology readiness level (TRL) of the technology, which is based on a range of 1 to 9, with 9 representing the most mature technology. The TRL assessment was determined following an extensive literature review through consideration of the most promising processes including estimation of the potential application of the technology for CO_2 utilization. Although supported by literature sources, the authors do, however, caution that the initial evaluation was limited in a number of cases due to a lack of data pertaining to the estimation of CO_2 reuse potential and, consequently, the TRL assessments should be considered as tentative. The analysis identified that the industrial processes with the highest TRL of 9 are bauxite residue carbonation, horticulture production and urea production. The processes with an estimated TRL of 7–8 are concrete curing of concrete blocks, mineral carbonation and lignin production. The processes with a slightly lower TRL of 7 are methanol production, polyurethane production and polycarbonate production. This analysis indicates which technologies may be potentially employed in the nearer term (i.e., those with the highest TRL) and those technologies that may require further development in order to be deployed in the medium term (i.e., those with a lower TRL). Additionally the researchers illustrated a methodology for assessing the potential of CCU in several countries from Europe [58], including Germany, UK, France, Spain, Italy and Poland. Industrial production underpinning this potential was evaluated through use of the Prodcom Database developed by Eurostat. This database includes statistics arising from the production of manufactured goods as well as mining, quarrying and manufacturing industries. The studies enabled calculation of the CO_2 potential for the industrial processes that were assessed for technology readiness and the results were as follows: concrete curing (22.5 Mtpa), horticulture production (22.0 Mtpa), lignin production (8.4 Mtpa), ethylene and propylene polymers (8.3 Mtpa), mineral carbonation (5.3 Mtpa), urea (3.9 Mtpa), methanol (2.0 Mtpa), polyurethane (0.3 Mtpa) and bauxite residue carbonation (0.2 Mtpa).

On the matter of the energy requirements for capturing and processing CO_2, Farmer and Doherty [59] have calculated both the absolute minimum energy penalty and efficiency-adjusted energy penalty for each of five CO_2 emission reduction technologies, which are as follows, respectively:

- Carbon capture and sequestration from point sources (CCS): 15.2% and 32.2%.
- Carbon capture and sequestration from air (CCSA): 20.0% and 46.6%.
- Carbon capture and utilization as structural materials (CCUSM): 8.2% and 9.2%.
- Steam methane reforming with carbon capture and sequestration (SMR + CCS): 11.3% and 20.0%.
- Methane pyrolysis (MP): 45.0% and 45.0%

The analysis is based on the relevant EPA (Environmental Protection Agency) coal emissions factors. Furthermore, it should be emphasized that the energy penalty for CCS will be dependent on the source of the carbon dioxide and the particular facility, such as a coal plant, natural gas plant, cement plant, steel plant or ethanol plant. The analysis reported in these studies is based on the average US coal fired power plant efficiency and includes corresponding assumptions, such as the EPA emissions factor for coal used in the US electric power sector being 95.52 kg of CO_2 per million BTU (British thermal unit) of generated heat. It should be further noted that the energy requirements for CCU will include both electrical energy as well as heat energy inputs as part of the corresponding industrial processes [59].

The energy penalties (EP) were calculated by the researchers [59] according to classical thermodynamics and based on minimum work calculations, where the absolute minimum EP is equivalent to the thermodynamically defined minimum energy costs for each industrial process and the more realistic (efficiency-adjusted) EP accommodates higher costs associated with additional operating and capital costs for the industrial processes. In this context, the EP can be considered as a convenient construct to examine the energy related costs of the CO_2 capture process, where the EP is defined as "the primary energy required to drive the CCS of a mole of CO_2 divided by the primary energy available from the combustion of the quantity of fuel that generates a mole of CO_2" ([59], p. 567). Consequently, EP can be considered as the fraction of the primary energy of a power generation fuel, which would need to be directed towards the CO_2 capture process. This analysis highlights that carbon capture and utilization as structural materials (CCUSM) compares favorably with the other CO_2 emission reduction technologies, where it has the lowest absolute minimum EP of 8.2% (*cf.* $\overline{x}$ = 20.0%, SD = 14.7%) and lowest efficiency-adjusted EP of 9.2% (*cf.* $\overline{x}$ = 30.6%, SD = 16.1%). Conversely, carbon capture and sequestration from point sources (CCS) has an absolute minimum EP of 15.2% and an efficiency-adjusted EP of 32.2%. The analysis is considered as indicative since the authors identify that the real processes will likely have higher energy-related costs in addition to there being further operational and capital costs linked to the corresponding industrial facilities. The calculations allow the different technologies to be compared and contrasted alongside conventional energy infrastructure through a process of identifying the lower bounds for the costs of technology implementation according to an equivalent energy basis. Moreover, the authors state that retrofitting CCS technology to current operational US coal fired power plants would potentially increase production costs by at least 50%. This assessment was based on energy costs alone as well as the assumption that ultimately the direct air capture process would be significantly more costly than CCS from a point source. Nevertheless there continues to be a need to reduce the energy penalties for all the different derivatives of carbon capture in order for the technologies to be commercialized on an industrial scale. Indeed, recent research has highlighted the scope for a zero-energy penalty CCU system, which is based on liquid fuel and power cogeneration via a process of chemical looping combustion [60].

4.3. Trend Estimation of CO_2 Usage

A key determinant for the ultimate success of deploying CCU technologies is the expected level of CO_2 usage, which would potentially be supplied through adoption of CCU. In this regard, the International Energy Agency [44] estimated both growth in global CO_2 demand over the years 2000–2025 (see Figure 7) as well as the breakdown of types of demand in 2015 (see Figure 8). It should be noted that Figure 7 includes projected figures for future global CO_2 demand for 2020 (250 Mt/yr of CO_2) and 2025 (272 Mt/yr of CO_2), which are given according to an average year-on-year growth rate of 1.7%.

Trend estimation based on linear regression (R^2 = 0.9964) provides an estimate of global CO_2 demand by the year 2030 to be ca. 300 Mt/yr of CO_2. According to this analysis, it can be observed that the global CO_2 demand is expected to increase at a steady rate over the next decade. It should be noted that in order to calculate an acceptable R^2 value (i.e., close to 1.0), the regression was limited to being linear and not exponential—with the latter corresponding to a much higher rate of growth. Nevertheless this analysis identifies the positive slope of the demand growth, where demand is expected to approximately double from the year 2000 (*ca.* 150 Mt/yr of CO_2) to 2030 (*ca.* 300 Mt/yr of CO_2), which underpins the scope for CCU to be deployed in order to meet this growth in demand. As identified in Figure 8, this growth in consumption of CO_2 (based on data from the IEA) is envisaged to be largely precipitated through adoption of enhanced oil recovery combined with the on-site demand for urea production.

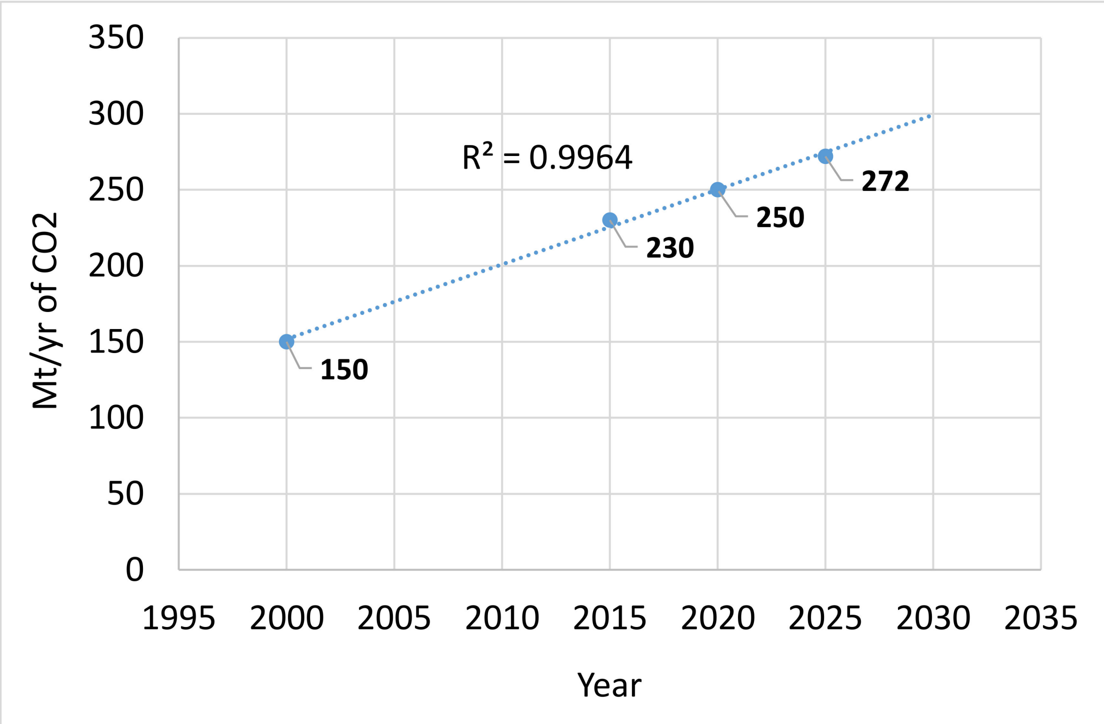

Figure 7. Growth in global CO_2 demand, source of data: IEA [44].

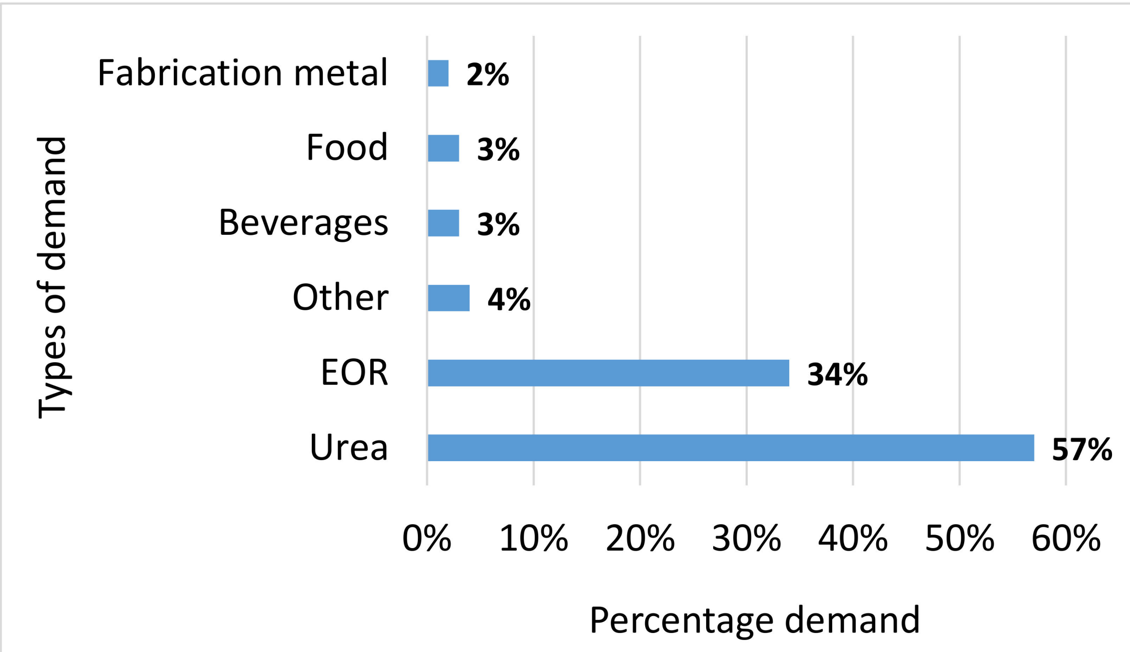

Figure 8. Breakdown of global CO_2 demand in 2015 according to type, source of data: IEA [44].

Since urea production is viewed as a major area of use of CO_2, it is useful to consider growth in global production capacity of this material, which is also known as carbamide (see Figure 9, sources of data: Aresta et al. [61]; Statista [62]). According to linear regression analysis (R^2 = 0.9191), it can be observed that the global production capacity of urea is envisaged to increase to approximately 300 million metric tons by the year 2030—which further underpins the scope for CCU to be deployed. A note of caution is required in regard to this analysis due to the limited availability of data points over the timeframe up to 2030 and therefore this analysis should be considered as tentative. However, the R^2 value being above 0.9 can be viewed as acceptable for the regression, which, despite not being non-linear (e.g., exponential), does nevertheless highlight the positive slope and growth in demand in annual global urea production. It should be further highlighted that existing urea production is undertaken in conjunction with the production of ammonia through capture of carbon dioxide from ammonia plants and that urea is mainly used in fertilizers and polymer synthesis [63]. In the case of fertilizer use, carbon dioxide is still released into the atmosphere and so there is no net benefit in terms of mitigating GHG emissions, although there are potential benefits from a circular economy perspective in terms of the reuse of carbon dioxide currently emitted from power plants where it could be utilized in urea production. Researchers continue to investigate urea production in the context of CCU and this includes, for instance, exploring the synthesis of urea at atmospheric pressure by negative corona discharge in gas phase [64] as well as the combustion of oxy-fuel in order to enable synthesis of urea and ammonia [65].

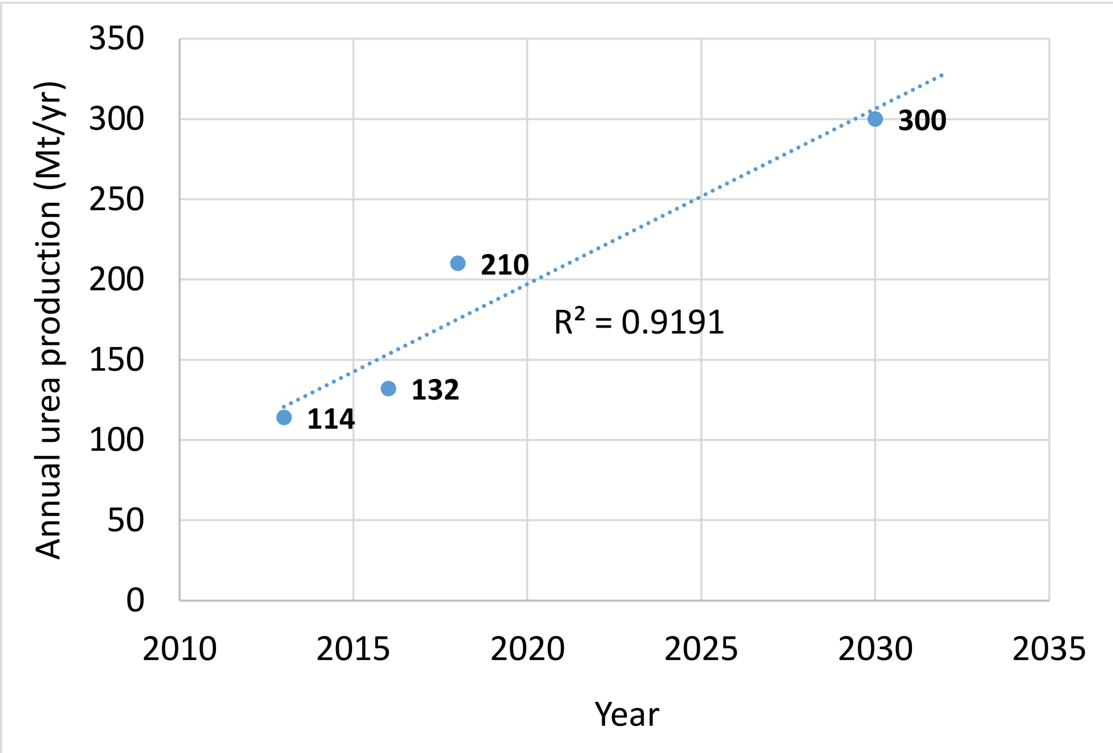

Figure 9. Growth in global urea production per annum, sources of data: Aresta et al. [61]; Statista [62].

Since it is envisaged that EOR will account for a continued significant component of global CO_2 usage, Figure 10 (source of data: McGlade et al. [66]) provides the growth in global EOR levels. According to exponential regression analysis (R^2 = 0.9923), it is estimated that global EOR levels will increase from 0.55 mb/d (thousand barrels per day) in 2020 to around 1.64 mb/d in 2040, although it can be observed that much of this growth occurs from 2025 onwards, where at this point it is estimated to be 0.81 mb/d. Nevertheless the non-linearity of the curve highlights the projected high level of growth in global EOR levels and according to an acceptable R^2 value. However, a note of caution is needed on this matter, since EOR is used to increase the production of crude oil, which is a carbon rich fuel with 93% of the carbon in petroleum being refined into combustible products that are ultimately emitted into the atmosphere [67]. Therefore, it is essential that the overall life cycle implications of EOR are properly investigated in order for the benefits of CO_2 sequestration to be adequately understood.

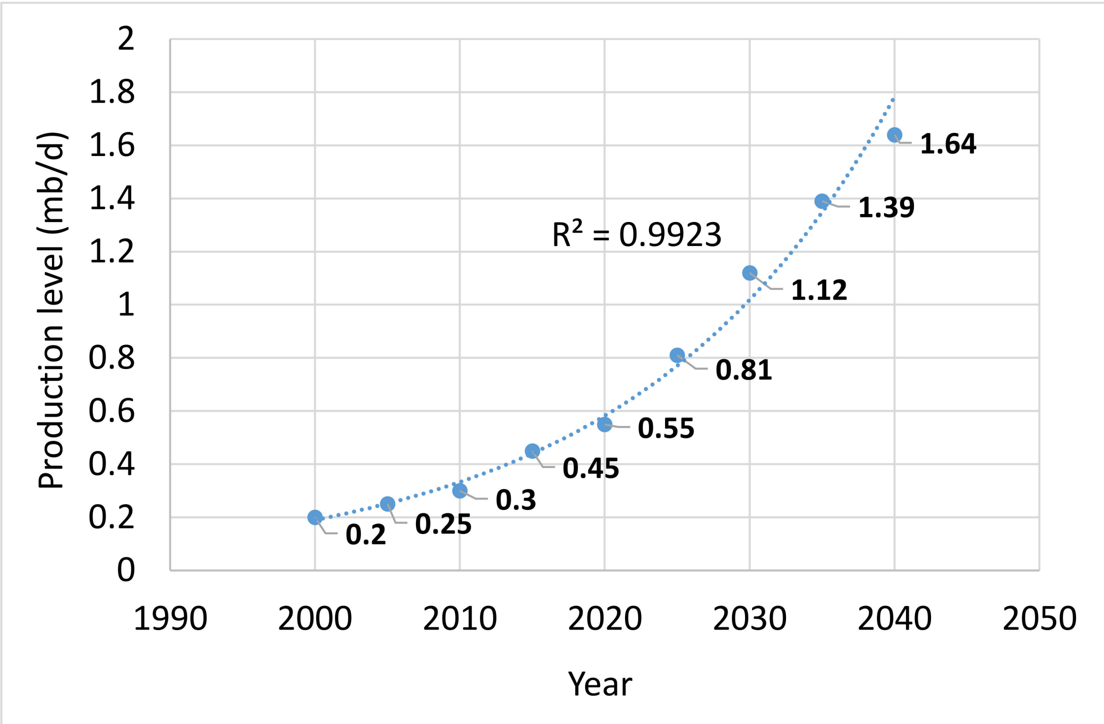

Figure 10. Growth in global enhanced oil recovery (EOR) levels, source of data: McGlade et al. [66].

4.4. Evaluation of Methane and Methanol Production

Various researchers have explored the potential scope to convert carbon dioxide to methane and this includes, for instance, CO_2 utilization via dry reforming of methane for the eventual purpose of enabling production of dimethyl ether [68], and the production of renewable methane from biomass via a CCU process [69]. Furthermore, the utilization of both carbon dioxide (CO_2) and methane (CH_4) has been explored by Fan et al. [70], where both gases were used as reactants and can therefore be a potential route to remove the gases from the atmosphere or from industrial processes. In this case, dry reforming of CH_4 (DRM) can be used as a suitable process to convert CH_4 and CO_2 to syngas, which is a raw material used for the eventual production of liquid fuels.

In regard to production of both methane and methanol, a CO_2-based production process has been modelled by Hoppe et al. [71]. This system also depicts how polyoxymethylene (POM) as an eventual polymeric material can be produced. In the conventional process, natural gas is converted to methane, which is further converted to methanol and then subject to polymerisation to POM. In the CO_2-based production process, CO_2 is captured from either raw biogas, waste gases from a cement plant, or from flue gases of a waste incineration plant (i.e., via a renewable channel). The CO_2 can then be converted into either methanol, or combined with H_2 (produced via wind energy power generation coupled with electrolysis of water) to produce methane. Thereafter, the methane is converted to POM via the intermediate synthesis gas and methanol mixture.

The modelling of different scenarios by the researchers Hoppe et al. [71] in Germany highlighted that carbon dioxide-based production processes are viewed as not being competitive when compared to more conventional production methods, which is mainly a consequence of high electricity generation costs and high investment costs for electrolysis as well as an unsupportive regulatory environment in regard to energy supply from the German grid. The analysis also revealed that production of a CO_2-based polymeric material will potentially exhibit a higher level of economic competitiveness when compared to the CO_2-based platform chemicals. Moreover, it was further postulated that several factors may potentially support more favorable economic conditions for the production of CO_2-based products in the coming years, namely a decrease in production costs of CO_2-based chemicals, enhancement of the regulatory framework to support technology commercialization as well as relatively low price levels for the renewable energy supply. This highlights the future potential for CO_2-based products arising from CCU.

Methanol (CH_3OH) is an important raw chemical material with approximately 85% used in the chemical industry as a chemical feedstock or as a solvent for synthesis and around 15% is used as a fuel source [72]. In terms of chemical conversion, methanol is used in the industrial production of formaldehyde (35%), methyl tert-butyl ether (MTBE) (25%), and acetic acid (9%). Methanol can be converted into synthetic fuels and it can also be added to existing fuels, such as gasoline, as part of a fuel mixture. Upon conversion to formaldehyde, this material is used as part of the production of various polymers. The typical production of methanol involves the mixing of natural gas with steam, which is heated and passed over an appropriate catalyst in a steam reformer [73]. The resulting gas and steam mixture is further converted into synthesis gas, which is then pressurized and converted into methanol via catalytic reaction that is finally distilled to give pure methanol. Methanol can be prepared from various sources including natural gas, coal, biomass, landfill gas and also from emissions of CO_2.

In the case of CO_2 utilization in methanol synthesis, which is coupled with a range of syngas production technologies [74]. This work evaluated a possible "methanol economy" through the capture of CO_2, which is co-fed with natural gas in order to produce syngas that is suitable for methanol (MeOH) synthesis; this was directed towards comparing two key aspects, which were CO_2 emission intensity as well as the extent of methane (CH_4) reliance. The findings of the economic scenario analysis identified the most promising production technology involved dry methane reforming (DMR with H_2 addition), which outperformed other scenarios in regard to CO_2 emission intensity as well as methane reliance. Modelling of the carbon dioxide integration in a natural gas-based methanol synthesis plant has also been conducted by Milani et al. [72], where the reformer product (i.e., syngas) is combined with a high-concentration carbon dioxide flow derived from the power plant carbon capture (PCC) process. The studies identified that this form of process adoption could potentially reduce methane uptake by 25.6% and lead to a decrease in the combined CO_2 emissions for both the power-plant and methanol-plant by 21.9%, thereby underscoring utility of this approach. Other approaches for methanol synthesis via catalytic processes are also under development, for example, the catalytic synthesis of methanol from CO_2 over copper in the presence of zinc and under hydrothermal conditions was investigated by Huo et al. [75] (2012). As with other areas of CCU, the production of methane and methanol from CO_2 sources as well as the resulting scope for polymerisation is an active area of research, where different technologies are being modelled and evaluated for eventual commercialization with industry [76].

5. CCU Value Chain Analysis

Value chain analysis [77] is a useful mechanism to map the key characteristics and areas of value for a given process, business or industrial sector. Indeed, value chain analysis has been applied to different energy sector applications, for example, examination of energy technology dependence in regard to geothermal power in the European Union [78], and supply chain analysis of mixed biomass feedstock supply systems for application in lignocellulosic sugar production [79]. Furthermore, Hasan et al. [80] developed a multi-scale framework for carbon dioxide capture, utilization, and sequestration (CCUS and CCU). Therefore, through a process of inductive reasoning that enables generalization of findings from analysis of specific instantiations, it is possible to synthesize a value chain for development of CCU technologies (see Figure 11).

This value chain for development of CCU technologies has a number of main features. The main part of the chain is based on the source, capture, transport and utilization stages (or links) in the chain, where each stage of the chain identifies key areas that are under investigation or require further R&D to be undertaken. The value chain is accompanied by a series of cross-cutting themes that represent different perspectives of CCU (namely industrial, governmental and societal) as well as the ultimate objective of CCU to reduce CO_2 levels and mitigate the impact of climate change. It is envisaged that the value chain can be used to help inform future research trajectories on CCU and assist industry to evaluate CCU as part of the technology development and commercialization process.

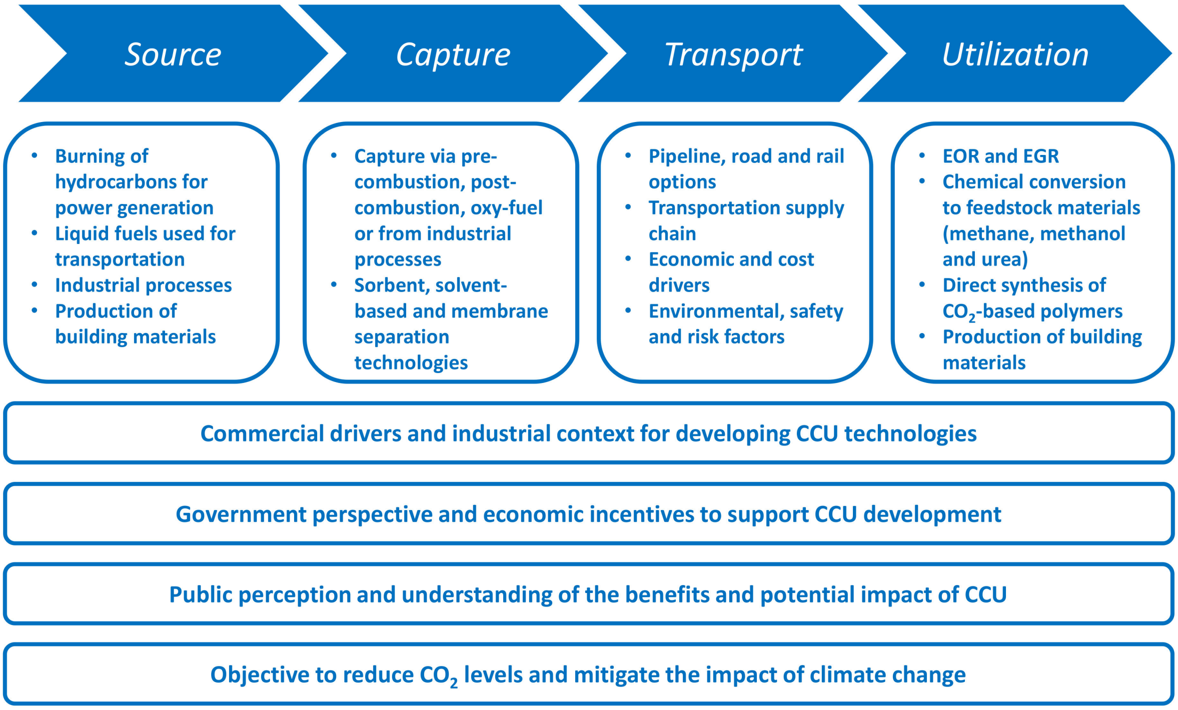

Figure 11. Value chain for development of CCU technologies.

6. Conclusions and Future Work

This article has provided a critical analysis and evaluation of the technology pathways for carbon capture and utilization (CCU), which has focused on the key areas to be addressed in order for CCU technologies to be commercialized for industrial application. CCU can be considered as part of a portfolio of options alongside CCS and renewable energy technologies that are available to policy makers and industry in regard to reducing CO_2 levels according to a sustainable circular economy solution. However, there remain a number of technological, economic and environmental challenges as well as regulatory and public perception issues that need to be addressed in order for CCU (and CCS) to be implemented on an industrial scale. Nevertheless, this technology provides significant opportunities and scope to not only reduce CO_2 levels but also stimulate innovation and commercial development as well as new supply chain configurations focused on the utilization of waste CO_2 for various industrial applications. The value chain developed in the research study can help to guide industrial development of CCU technologies across the source, capture, transport and utilization stages of the chain and through accommodating the needs of the various stakeholders.

The research study focused on the use of the critical analysis and evaluation method and the specific findings can be summarized as follows:

(1) CCU is an emerging area of research as identified by the bibliographic study and the analysis suggests that despite the various challenges, CCU will continue to grow as an area of interest for researchers, industry and governmental organizations. The main subject areas where CCU has been investigated include energy, environmental science, materials science, chemical engineering and general engineering as well as several other areas to a lesser extent. Indeed, since there are many technological, economic and environmental challenges associated with implementing CCU, adoption of a multidisciplinary research perspective is encouraged and will be essential if the industrial development challenges identified in this article are to be surmounted.

(2) There are a range of carbon dioxide utilization processes currently available, although not all are fully developed for industrial application. EOR is an existing practice (i.e., high TRL) available in certain cases to significantly increase the level of production from oil wells that are depleted via traditional extraction with the benefit of sequestering CO_2 in geological rock formations on an effectively permanent basis, although there is the overall net effect on carbon emissions to be considered in regard to higher levels of petroleum products eventually arising from EOR. Chemical conversion to feedstock materials including methane and methanol have a relatively high TRL and are active areas of development alongside existing production of urea (although this will still likely lead to carbon emissions). Additional synthetic pathways to polymers and rubbers represent an emerging area of CCU, which is currently at a low TRL but has significant potential for the future. Other CCU processes (such as mineralization as well as application to the production of building materials, e.g., concrete) are less developed and have a lower TRL.

(3) The global level of demand for CO_2 is expected to continue to grow in a steady and linear fashion, with much of this current demand being driven by EOR and urea production, although there is scope for growth in chemical conversion of CO_2 to other materials, as stated above. There is the potential for the level of EOR to grow at an exponential rate, although this will be impacted by the demand for oil and gas as well as the parallel development, maturity and lowering of costs for renewable forms of energy. However, the long term environmental impact of EOR needs to be properly evaluated due to the higher levels of oil and gas production and resulting carbon emissions that eventually arise from adoption of this process.

(4) The production of methane and methanol as well as further derivative materials represents an area of active development where there is scope to deploy new technologies to facilitate the transition to greater levels of CCU. The development of new CCU-enabled economies and business models for CO_2-based products, including methane, methanol as well as polymers such as polyoxymethylene (via syngas), is an area subject for further investigation and technological commercialization. In cases where there are favorable changes to the regulatory environment and a supporting economic case, such production routes are likely to become more viable and subject to accelerated industrial development.

In regard to future work, it is suggested that further modelling is conducted on the specific conditions required to support commercial development of the different forms of CCU and corresponding technological options. Consequently, there is a need for an improved understanding of the environmental implications of the various CCU technological development areas, including life cycle assessment as well as validation of circular economy-based business models for CCU. Thereafter, international comparative studies need to be conducted to help researchers target their efforts, and to inform policy-makers and industrial companies in order to focus investment and prioritise support for the development of leading CCU candidates. Further research is also suggested on investigating the economic, technological and environmental conditions required to support CCU adoption, including the required supply chains and industrial infrastructure, such as CO_2 capture and transport capabilities as well as engineering plants for processing and converting CO_2.

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