

Article

# Experimental Development of Calcium Looping Carbon Capture Processes: An Overview of Opportunities and Challenges

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**Abstract:** Global warming might be mitigated if emissions were interrupted through carbon capture technologies, as there is a significant amount of comprehensive studies on them. An outline of the main gaps and trends of a technology is critical for further development. In this context, this study provides an overview of calcium looping carbon capture processes that have proven their potential and commercial viability. A bibliometric analysis is conducted on both Scopus and Web of Science database by seeking the keywords “calcium looping”, “co2 capture”, and “fluidized bed” in titles, abstracts, and keywords. Word selection was based on a list of relevant papers on the topic. These items of data have been processed and analyzed based on the number of publications and citations by emphasizing recent publication evolution, journal influence, the use of specific keywords, and co-citation. Results reveal that the European Union (EU) leads the rankings on the topic, followed by Canada. Keyword choice might have affected the number of citations. Recent studies used limestone as a sorbent and a dual fluidized bed reactor with a calciner or resistance depending on its size. Most studies are focused on technology scale-up. Although scale-up seems to be a priority, multiple studies are designed to assess the effect of steam generation and SO<sub>2</sub> on the process.

**Keywords:** calcium looping; carbon capture; fluidized bed; bibliometric analysis; scientific gaps



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## 1. Introduction

According to the Intergovernmental Panel on Climate Change (IPCC), historical cumulative net CO<sub>2</sub> emissions from the years 1850 to 2019 were 2400 ± 40 GtCO<sub>2</sub>, and 42% of this increase occurred between 1990 and 2019 [1]. Although damage not only seems to be inevitable for every scenario after the year 2100, even by considering that there might be a cessation of net anthropogenic CO<sub>2</sub> emissions, high net CO<sub>2</sub> removal from the atmosphere may prevent irreversible multi-century damage in the long term [2].

Carbon capture technologies are developed with the aim of mitigating climate change. This process consist of CO<sub>2</sub> removal from a given industrial process that is aimed to be subsequently stored or reused [3], all of which is bound by the Paris Agreement. Many strategies have been developed and reported, such as post-combustion capture [4], pre-combustion capture [5], absorption techniques [6], the electroreduction of CO<sub>2</sub> [7–10], and the use of membranes [11]. CaL, a derivative form of chemical looping from post-combustion for carbon capture, has demonstrated scale-up potential and commercial viability in the literature [12–14].

The CaL process consists of a reversible carbonation–calcination reaction of a CaO-based sorbent [15] by mostly using limestone. Limestone is an abundant low-cost material

that is suitable for an economically viable process. Natural limestone is largely utilized, even though some authors have proposed using pellets and materials that are modified in the lab [16–22]. The process requires a dual fluidized bed reactor to produce a cyclic capture and release of CO<sub>2</sub>. The CO<sub>2</sub> contained in flue gas is captured by a carbonator; then, CaO forms calcium carbonate (CaCO<sub>3</sub>) according to Equation (1).



Natural and synthetic sorbents are of paramount importance to ensure the high efficiency of CaL processes. However, the sorbent's stability should be demonstrated after several calcination/carbonation cycles. For these evaluations, CO<sub>2</sub> capture capacity, fast CO<sub>2</sub> absorption kinetics, strong mechanical strength, and low friction rate should be taken into account [23]. Although dolomite has a low capture capacity at the beginning of the process, it has greater stability, which leads to good overall performance in further carbonation/calcination cycles [24]. This occurs on account of the porosity of the sorbents, which remain unchanged during the process due to unreacted MgO, which has a higher melting point than limestone, thus minimizing the sintering mechanism [24]. Although the CaL process is promising, research has shown that the reversibility of carbonation/calcination reactions is far from ideal. Its limitations usually refer to a reduction in carrying capacity, the sintering of sorbents, competition with the sulphation reaction, and the attrition of particles in reactors.

Doping agents such as CaBr<sub>2</sub>, KMnO<sub>4</sub>, Mn(NO<sub>3</sub>)<sub>2</sub>, MnCO<sub>3</sub>, NaCl, Na<sub>2</sub>CO<sub>3</sub>, KCl, K<sub>2</sub>CO<sub>3</sub>, and sea salt have been used for improving natural limestones and dolomites. At first, this step reduces the initial capture capacity, but a higher structural stability is achieved, with an approximately 5% increase in the number of cycles [23]. This is achieved through a reduction in the decarbonation temperature and the creation of vacancies during decomposition by alkali carbonates, which leads to an increase in ion diffusion and the acceleration of CaO sintering [25]. According to Han et al. [25] the long-term stability of the reactor and components in CaL heat storage is still unclear, and further research and trials are needed. An increased cycle number leads to a decrease in the capacity of sorbents to capture and release CO<sub>2</sub>. This phenomena is known as 'capture capacity' [26]. A decrease in capture capacity is normally attributed to the loss of sorbent porosity [27]. Limestone sintering refers to a change in the shape and size of pores, as well as the grain growth during heating [28]. This sintering process is exacerbated by prolonged exposure to high temperatures, and it is reinforced by the presence of CO<sub>2</sub> and/or steam [27].

In addition to reacting with CO<sub>2</sub>, CaO can also react with sulfur-containing compounds, such as SO<sub>2</sub> (in combustors) or H<sub>2</sub>S (in gasifiers). According to Anthony and Granatstein [29], at temperatures lower than 700 °C and under oxidizing conditions (the same calcinator conditions), limestones are more likely to form CaSO<sub>4</sub> than CaSO<sub>3</sub>. The friction of CaO is attributed to its friable structure [30]. Scala and Salatino [31] characterized sorbent attrition in fluidized bed reactors based on the size of broken fragments and the associated breakage mechanism, which involves the following: primary fragmentation (thermal and mechanical stresses) and secondary fragmentation (particle collisions and particle wall). Sorbent abrasion, however, generates fine particles, which are typically elutriated.

In CaL processes, different variables can affect the efficiency of CO<sub>2</sub> capture, which can be related to the characteristics of the raw material, reaction conditions, and the type of reactors being used in the process. Calcination and carbonation temperatures are considered to be the most relevant parameters in CaL processes [32]. The loss of sorbent reactivity is influenced by attrition, and such mechanism includes fragmentation (primary and secondary) and abrasion [24,33]. Sorbent sintering also occurs based on the number of heating and cooling cycles in CaL processes [23] and through an increase in the partial pressure of steam and CO<sub>2</sub> [24]. The choice of reactor type, residence time, and reaction atmosphere are also variables affecting CaL processes. Thus, it is often necessary to carry out numerous experiments to identify the variables affecting the process.

In order to minimize the number of experiments and determine influencing factors, the design of experiments (DoE) can be applied [34]. DoE methods have been widely used [35,36] as a method to evaluate the effects of numerous independent factors and their interactions on the response variable of interest as an optimization condition [37]. They have also been used to reduce essential experimental data to achieve the best operating conditions for an optimal response in various processes [38], in addition to substantially improving reaction conditions and the performance of chemical processes [39], which can be applied in processes related to carbon capture.

Fluidized bed reactors are very commonly used in gas–solid reactions due to excellent gas–solid contact on the bed, efficient gas particle heat and mass transfer, and high bed wall and internal heat transfer coefficients [40]. Calcium-looping applications stand out on account of being associated with a higher conversion performance [13,41]. Researchers have adopted experimental configurations with two independent reactors on an industrial scale [12,13,15,42–44]. Nonetheless, many researches have proposed to enhance its viability through the use of solar power [41], waste-derived fuels [13], CaO-based lab-fabricated pellets with enhanced properties [45], and high-capacity carbonators [15]. However, such technologies are still being scaled up [12].

H<sub>2</sub>O and SO<sub>2</sub> associated with the carbon capture capacity of limestone was also investigated [46]. The authors found that SO<sub>2</sub> was damaging with regard to CO<sub>2</sub> capture performance, while steam positively affected it. Thus, it was evidenced that the beneficial effects of steam outweigh the negative effects of SO<sub>2</sub> at low SO<sub>2</sub> concentrations. Although the benefits of adding steam to the process are acknowledged, its application has not been fully explored yet [47]. Other hydration techniques are currently being suggested [48].

Moreover, it is vital to thoroughly understand trends to scale up the technology more efficiently and sustainably. Understanding how concepts differ in their effects on scale-up technological progress, its major obstacles to advancement, and how specific nations have invested in this technology can assist in better comprehending how these factors affect future decisions.

Thus, this article aims to perform a bibliometric analysis of experimental fluidized bed reactor configurations in CaL operations. The same search was performed on the Scopus and Web of Science databases to compile our database. Data were cleansed, analyzed, and processed based on eligibility criteria.

The findings show how the technology has evolved according to various authors' contributions, as well as how paper production and citations have changed over time. The main scientific gaps are outlined, along with suggestions for a better utilization of keywords in order to reach a greater number of citations.

## 2. Methods

### 2.1. Database Formation

To start compiling the database, the topic should be well outlined. In this context, the database search should reveal works related to experimental data and setups for CaL utilization. A total of ten manuscripts from relevant paper references were selected, while a manual check was conducted to ensure that works had experimental data, an experimental apparatus description, a material description and that they were conducted in the context of CaL processes using a fluidized bed reactor. The previous requirement was used to lay down the following eligibility criteria:

- It should not be classified as a conference, abstract, or be under consolidation;
- It must be focused on calcium oxide (CaO) and limestone (CaCO<sub>3</sub>), regardless of whether it has undergone a procedure to enhance its properties or not;
- The research scope must be within experimental carbon capture using fluidized beds, which should be bubbling or circulating;
- The primary focus of the paper must be the development, specific details, phenomena, and limitations of the technique used for CaL carbon capture using fluidized bed reactors;

- In order to validate experiments and analyses, CaO and CaCO<sub>3</sub> conversion processes must be observed in at least one complete cycle.

They were used to reveal that the whole database has been filtered in accordance with such criteria.

Keywords were collected from ten papers and listed in order of frequency. The most frequent keywords were “calcium looping”, “co2 capture”, and “fluidized bed” with eight occurrences each, followed by “attrition” with four occurrences and “limestone” with three. An additional 13 keywords were also found; however, they were disregarded since they were found only once on the list. The remaining keywords were “attrition” and “limestone”, which restricted the search only to natural limestone; therefore, they were also disregarded.

An online search for recent publications was performed on 14 February 2023 on the Scopus and Web of Science databases in an attempt to find the words “calcium looping”, “co2 capture” and “fluidized bed” in the title, abstract, or keywords of papers.

A total of 133 results from the Scopus database were obtained after inputting the selected keywords. Only final editions of research papers published in English were retrieved. Ultimately, 103 manuscripts remained, and 98 of them could be accessed. The results were manually narrowed down to the topic of ‘experimental CaL technique applied to fluidized bed’ using the eligibility criteria. Similarly to the process conducted in papers to establish criteria, database papers were downloaded, and the information present on the criteria was sought manually. Only papers containing all these items of information were selected. Based on these criteria, only 37 items satisfied the requirements. These papers were then organized by author names, title, journal title, author keywords, indexation keywords, abstract, addresses, references, number of citations, year of publication, DOI, database source, country, and affiliation on a data frame.

The same procedure described above for the Scopus database was used for the Web of Science database, and it yielded 71 results. A total of 65 items remained after applying the filters. The result of a comparison between the two databases found 19 duplicate articles, which were then removed from the list to result in 51 papers, but access was only granted to 49 of them. After applying eligibility criteria, only five papers remained. They were separated into a new data frame combining the Scopus and Web of Science databases. The final data frame comprised a total of 42 articles.

## 2.2. Data Analysis

Documents were extracted in a CSV (comma-separated values) format from the Scopus and Web of Science databases. Due to variances in the structure of both files, it was necessary to standardize data collection. The Scopus database was used as standard. Data columns inconsistent with the current analysis were removed. Then, documents were arranged according to their year of publication and included the following details: authors’ names, document title, source title, keywords, abstract, affiliations, and DOI (digital object identifier). The information provided was used to carry out a variety of bibliometric analyses. This paper regards that publication relevance entails a large number of citations; therefore, the year of publication served as a weighted indicator of significance. In this case, a study published four or ten years ago might have had a similar amount of citations, but, due to the time required to compile a paper, there was insufficient time for research published to receive citations.

This analysis assessed the relevance of articles using the number of citations as the criteria. A publication with a great number of citations is believed to provide a distinctive viewpoint on a subject or offer solutions to problems.

In order to classify authors, the citations must be attributed in a structured way. Citations were attributed through two steps. Firstly, for each paper, the authors were arranged, and the total number of paper citations was assigned to the first author. Secondly, after assigning the citation for every paper, the authors were organized in descending order by summing citations.

The dataframe assessment process made it possible to identify research trends, key results over the years, experimental setup dimensions, materials, sizing, and scientific gaps that might not have been filled yet. Additionally, an international overview of research development was performed by assessing the number of articles published by different nations and authors. Furthermore, a correlation between keywords used to create our database and those which generated a greater number of citations by database was examined.

### 2.3. Co-Citation Analysis Methodology

This analysis was restricted to the Scopus database (37 articles), since it was impossible to export citation information through the WoS database.

There was a list of references associated with every input. Citation information is composed of authors names, paper name, year, journal name, and page number. For this analysis, information was filtered using only authors' names. For every author cited, their name was attributed to each collaborator of an input paper.

Two new lists were compiled: one only with authors based on publication and citation and the other having a relation between publishing author and cited author, in which each author of a published paper received all author citations. Every link with equal names was then removed from the list.

A list containing only one author's publications and citations was vast and contained a large amount of non-critical information. It was found that many citations were not correlated with the remaining publishing authors, and the list was filtered so that only authors included on the lists of publications and citations were selected.

A list of unique authors was used as the central node of the analysis. In order to define the strength of peer cooperation, the number of times an author cited another was counted, and, the larger the number, the wider the bar and the greener the color of names was.

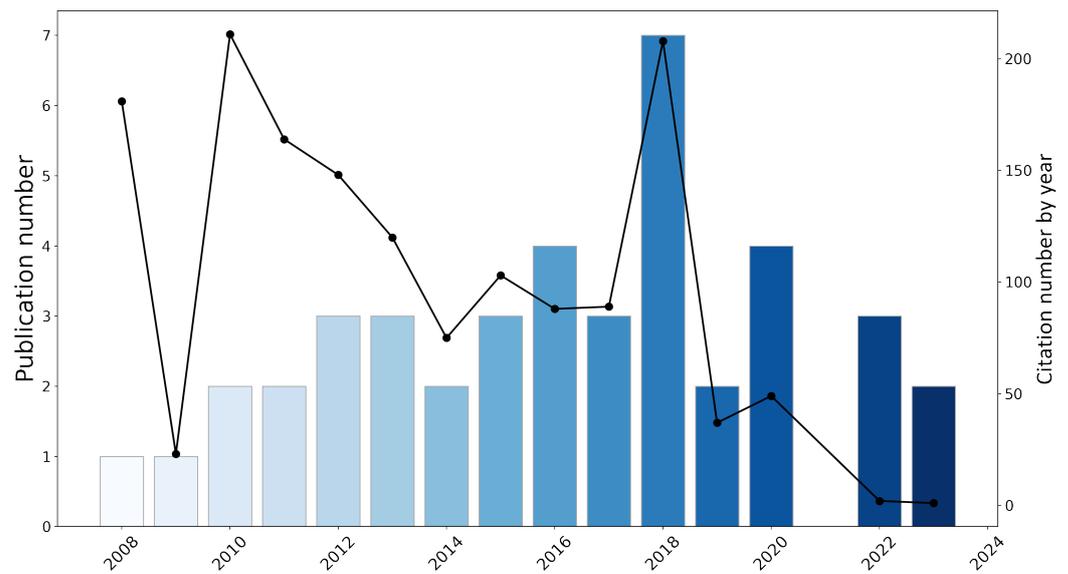
## 3. Results and Discussion

### 3.1. Analyses of Publication and Citation Tendencies

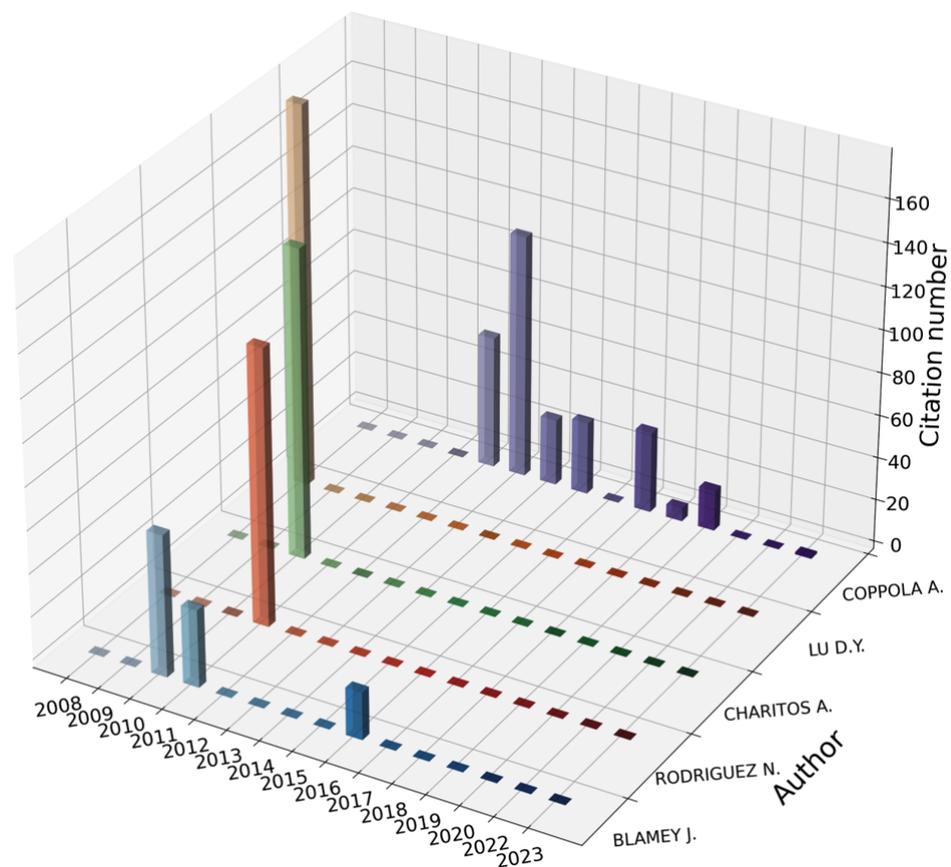
The number of documents and citations per year that reveal publication patterns are displayed in Figure 1. Three periods are the most relevant: 2008, 2010–2012, and 2018. A rising trend in the number of publications can be observed, starting from papers published in 2008. In the following years, there was a continuous increase in the number of documents published until 2013, after which the annual production remained constant until 2017. Compared to earlier years, 2018 showed a significant increase in the number of documents published, which might be related to the Paris Agreement being concluded in 2016. In 2019, there was a decline in production, which was expected, given the results of 2018. 2020 also had larger numbers of above-average documents, which indicated a steady growth in the amount of knowledge on the subject. There was a small number of articles published between 2021 and 2022, possibly due to the COVID-19 pandemic, which might have halted scientific research.

An examination of the most frequently cited authors was performed to more precisely indicate essential research in Figure 1. This analysis can assist in the discussion, once the author and research receiving the majority of a given year's citations can be identified. Figure 2 shows the five most cited authors from the data frame, along with their respective publications over the years.

Lu et al. [49] appeared first on Figure 2, and they introduced a dual fluidized bed reactor setup, with each bed being responsible for a single reaction in the CaL process. This investigation revealed recent research complications, such as the attrition of particles [12,46]. Moreover, the authors clearly stated that several processes were happening on the sorbent surface, which could be seen in other experimental setups, e.g., using fixed bed reactors. The number of gaps and the novelty of works might be a reason for greater interest in this specific technology. 2008 was the year during which there was the highest number of citations on the current analysis, with Lu et al. [49] being the only 2008 work.



**Figure 1.** Evolution of database citations and publications over the years.



**Figure 2.** Current data frame arranged from the most cited to the least authors.

Charitos et al. [50] investigated the effects of carbonator space-time, carbonator temperature, and the CaL ratio on a dual fluidized bed reactor plant. Its relevance might be related to parameter denomination and experimental measurement range. Carbon capture efficiency was firstly reported on the data frame as being 90%, and the experimental temperature ranges of maximum CO<sub>2</sub> capture efficiency were provided (600–660 °C). Carbon capture efficiency is related to the CaL ratio and space-time. Furthermore, the effects of sintering were presented and its aggravation with higher cycling, which also impacted CO<sub>2</sub>

carrying capacity, pore volume reduction, and a decrease in particle surface area. Finally, data on the dolomite attrition ratio were 2 wt%/h of total mass. This paper received about 69% of the 2010 total citations, which was quite likely considering the information and measurement reference.

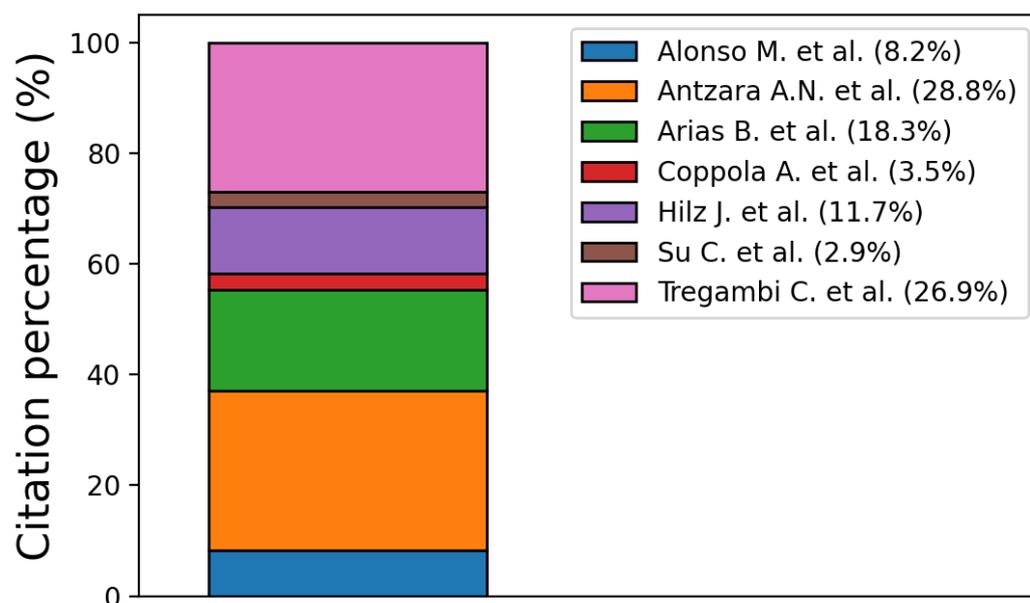
A paper published by Blamey et al. [51] had 31% of all citations in 2010. The authors investigated the hydration effect of a CO<sub>2</sub> sorbent during the CaL process using a fluidized bed reactor with the aim to mitigate sorbent inactiveness after a few cycles. A higher efficiency was found after a hydration pretreatment of the sorbent, although greater particle fragmentation was also observed due to the stronger sintering effect caused by cycles. Within the evaluation period, this is the very first work on using a pretreatment of sorbent hydration to reactivate it. In addition, they introduced new issues regarding sorbent hydration in a fluidized bed reactor.

A study carried out by Rodríguez et al. [52] received 78% of all citations in 2011, wherein they studied constant CaO flux on the carbonator, and their work was the first of its kind in our database. Due to deactivated particle accumulation inside the carbonator, it presented the cycling of active particles in order to ensure a constant and minimum quantity of activate material inside the fluidized bed reactor. Furthermore, the authors suggested that the efficiency of a sorbent with a larger number of cycles can be improved by keeping it on the fluidized bed for longer than the newer sorbent, thus enhancing carbon capture efficiency.

Coppola achieved a total of four publications between 2012 and 2013, among which Coppola et al. [30] and Coppola et al. [53] were the ones receiving the largest number of citations. Coppola et al. [30] was the first work performed by Coppola and collaborators to be found in our database. It offers an analysis of the effect of SO<sub>2</sub> on carbonation, which was evidenced to be present and harmful to the process [54–56]. Furthermore, the authors reported that SO<sub>2</sub> concentrations between 100 and 1800 ppm produced similar results, thereby demonstrating that the SO<sub>2</sub> in flue gas greatly reduced the sorbent CO<sub>2</sub> capture capacity. Additionally, a small increase in attrition was caused by SO<sub>2</sub> during particle carbonation. An interaction between the degree of calcium sintering and the sulphate layer properties generated on the sorbent surface regulated the propensity of limestone to undergo attrition. The SO<sub>2</sub> ratio found in fixed beds was also related to fluidized beds and elutriation results. Coppola et al. [53] verified the elutriation effect on six different types of limestone using a fluidized bed reactor. The study presents the effect of sintering during the first cycles and found that SO<sub>2</sub> affected the process and presented an average elutriation of 0.5%/h. There was a weak relation of SO<sub>2</sub> to elutriation, in addition to the fact the SO<sub>2</sub> effect on elutriation was introduced therein and is still under discussion recently [46].

2018 was the year that achieved the highest number of articles, and it also had a large number of citations. To better understand 2018 (the column presented in Figure 1), an in-depth analysis was conducted, which can be seen in Figure 3, so as to enhance visualization of each individual author's impact on a given year. Among the most cited papers, it can be seen that 28.8% (60) of the total citations were to Antzara et al. [18], 26.9% (56) were to Tregambi et al. [41], and 18.3% (38) were to Arias et al. [43]. Together, these authors were responsible for 74% (154) of the total citations in 2018.

Antzara et al. [18] investigated Zr-promoted CaO-based CO<sub>2</sub> sorbents under various conditions using a fluidized bed reactor with the aim to analyze the decay properties of a sorbent material. The proposed material showed superior properties compared to natural limestone in all aspects. Its larger number of citations might be due to the excellent performance of the proposed material. A hydration analysis was also conducted, and a conversion rate of 85% was found; thus, the capacity of this synthetic material was 5.6 times greater than natural limestone. Its higher performance allows a better use of this hybrid material in the future as the amount of material is reduced, and, consequently, its cost is also reduced due to higher efficiency.



**Figure 3.** Percentage distribution of author citations in 2018.

Tregambi et al. [41] used solar energy and a solar concentrator to heat up a CaL reactor. The authors reported a successful CaL cycle, although there was some temperature heterogeneity. Using solar power to conduct the heating might have been the reason for its large number of citations, given that it used clean energy to aid the process and reduce emission rise chances even further, in addition to enhancing the efficiency of CaL technology.

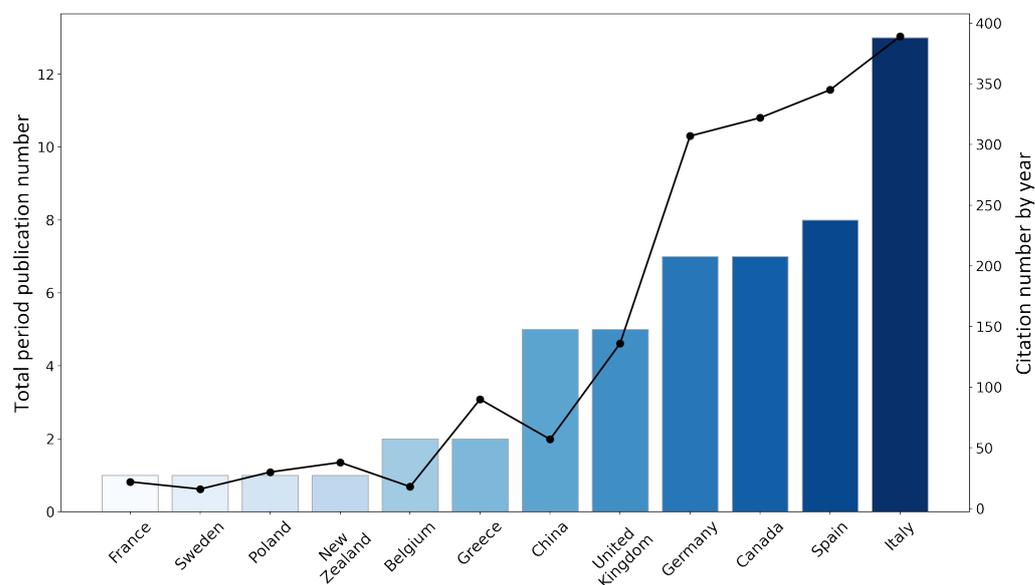
Arias et al. [43] investigated the feasibility of an ultra-rich oxygen condition on a calciner used in La Pereda CaL pilot plant. After a few modifications of their experimental setup, it was possible to perform tests using a max concentration of 75%O<sub>2</sub>. The results confirmed the efficacy of its operational condition, which was attributed to the endothermic nature of calcination and the large glow of solids circulating within the carbonator.

After an in-depth analysis of the most relevant years in Figure 1, it can be assumed that its high citation number might be related to the novelty of the experimental setup [41,49], the measurement methods and new experimental data [30,43,50,53], and the novel techniques and methods used to solve known problems [18,52]. However, the stage of technology development might have affected the way such information was perceived in the literature. Furthermore, once this data frame is restricted to experimental research investigation, the topics pointed out above might not be true for different areas.

### 3.1.1. Total Publication by Nation

Figure 4 illustrates the total number of papers published by nation within the years 2008–2023 that were compiled in our database. A total of 12 nations appeared on this list, including 8 members of the EU. The western hemisphere is represented by Canada, and the eastern one by China and New Zealand. The fact that many countries are not included in this list is possibly due to a lack of incentive for CC technologies. Another point worth mentioning is the absence of developing countries, which indicates a divergent global contribution to carbon emission mitigation technologies. The large number of EU publications might be due to specific investments. Recently, under project Horizon 2020, two projects have been focused on CaL technology [57,58]. They were developed from 2017 to 2021, which might have affected the number of publications in 2018. Both projects had a total cost of 14,213,254 EUR and were coordinated by research groups in Spain and Italy. In addition, both projects involved twelve countries, many of which are included in Figure 4. The relationship between nations' numbers of publications and their citation or paper relevance can also be observed in Figure 4. Italy also had the largest number of citations in our database, with a ratio (publication/paper) of 29.9. The lowest ratio was

represented by the nations having only one publication. The highest ratio belonged to Canada with 44.7. Canada has invested CAD 319 million in carbon capture, utilization, and storage technologies according to Natural Resources Canada [59]. Even though its funding is divided among different technologies, it seems as though a large sum of investment contributed to achieving more paper citations.



**Figure 4.** Data frame classifying countries according to their number of publications and total citations

### 3.1.2. Journal Analysis

Journals were been investigated based on their number of citations and publications. Although every journal had their cite score and impact factor, such parameters might differ for a specific topic as the one explored herein. Figure 5 presents the number of publications and total sum of citations from the data frame. It can be seen that powder technologies had the highest number of papers for this current topic, although it achieved a low number of citations (109), which suggests that this topic might not be in the scope of this specific journal. By observing the ratio between the amount of publications and the number of documents, the rankings changed as follows: the AICHE Journal (63.5) was ranked first, followed by the International Journal of Greenhouse Control (63.0), Fuel Processing Technology (46.7), the Chemical Engineering Journal (45.7), Energy and Fuels (45.0), Industrial and Engineering Chemistry Research (40.5), Proceedings of The Combustion Institute (29.5), Fuel (27.8), and Powder Technologies (18.2)

For this particular subject, in order to optimize exposition and citation, selecting one of the three journals achieving a higher ratio of publication number/number of documents seemed appropriate.

### 3.1.3. Keyword Analysis on Citation

Figure 6 shows the top five cited keywords, among which 'co2 capture' and 'calcium looping' were the first two keywords on the list. The most cited ones, representing the main subject of this analysis, were also used in the data frame. However, the third keyword, 'attrition', was used differently, as it is an intrinsic characteristic of the process. Even though 'carbonatation' appeared on the list as the main keyword on the topic of calcium looping, it was observed that keyword choice might have reached a higher number of citations to a given work if it was related to the main barriers and gaps found about it. Furthermore, associating the main subject with a keyword is essential to include the work in a given subject, but not addressing gaps and conclusions while selecting keywords could restrict views and citations. This result reveals that two or three keywords addressing the main subject were enough to delimit a topic, and all remaining keywords should be used to

highlight gaps to be filled and barriers to be faced. Nonetheless, an analysis of Figure 6 clearly evidences the fact that keyword use reached a maximum number of citations. Therefore, it is important to investigate other databases and themes so as to verify coverage of this statement, given that this result is only valid to this specific theme and data.

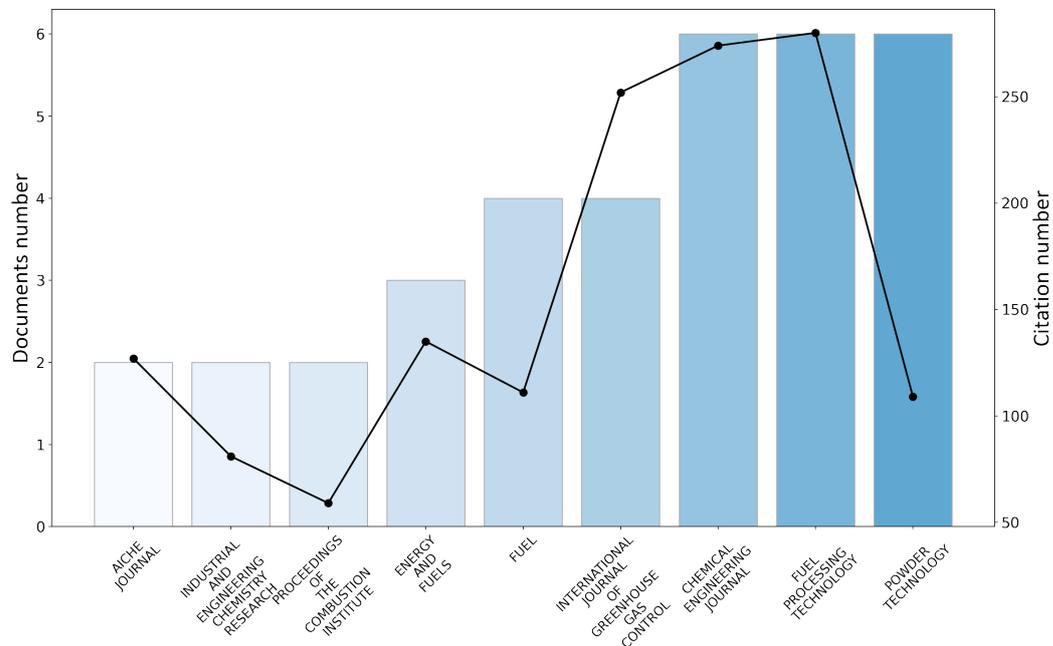


Figure 5. Most cited journals by number of documents and citation.

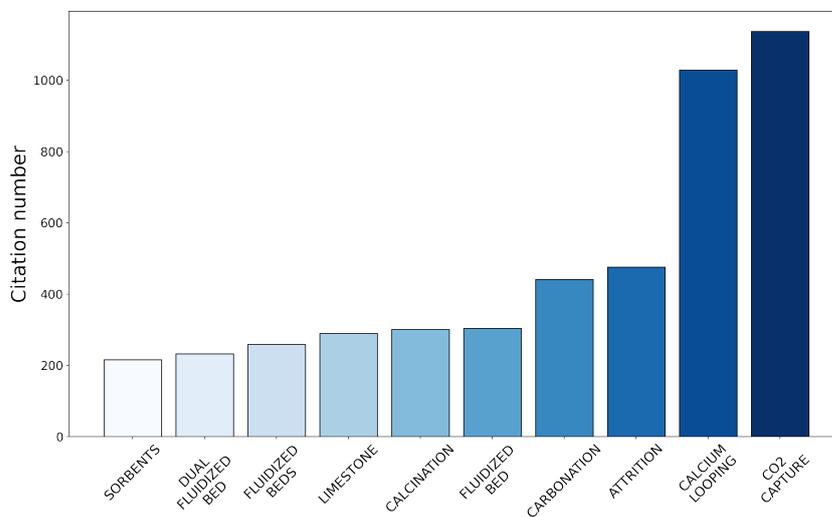


Figure 6. Five most-cited keywords.

### 3.1.4. Co-Citation Analysis

Figure 7 presents the analysis of citations between the data frame authors and their citations. Two main clusters of data can be observed: one of strong interconnected names and a peripheral one with many weak connections pairs. A central node (a node with many different connections) is understood as a diverse author with extensive cooperation with different group or individuals and a strong connection (wide dark green bar) entails a strong cooperation between the two names.

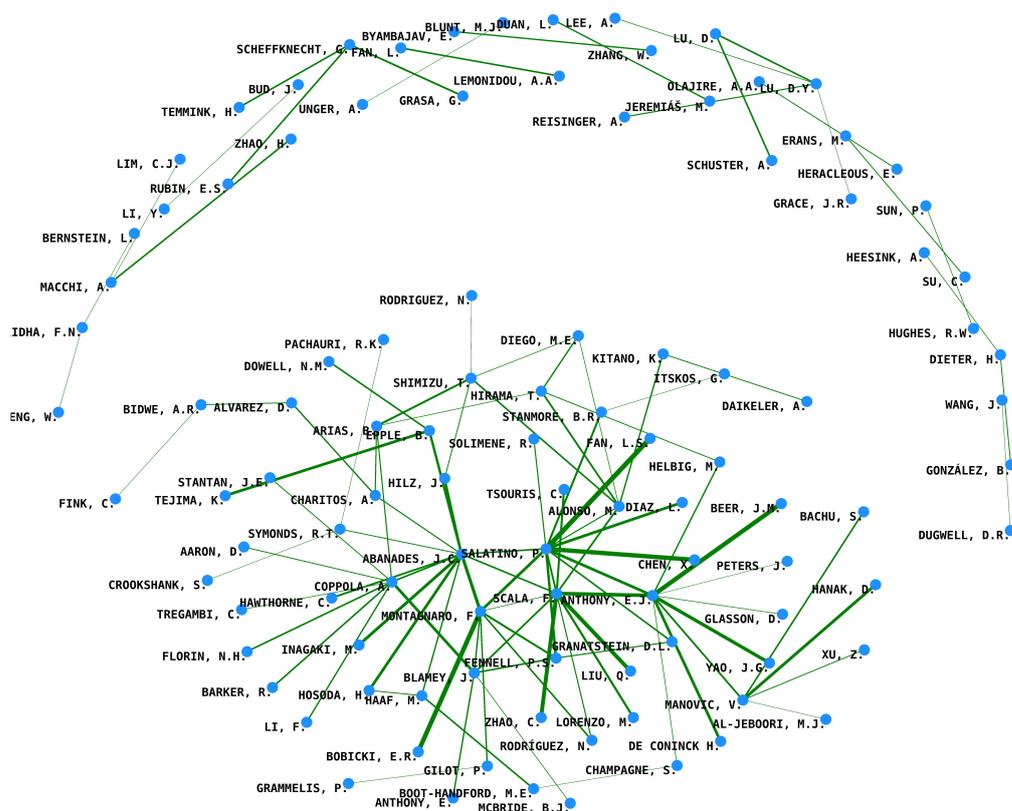


Figure 7. Data frame co-citation analysis.

The most-cited authors in Figure 2 (COPPOLA A., LU D.Y., CHARITOS A., RODRIGUEZ N., and BLAMEY J.) can be found at different places as central or peripheral nodes; nonetheless, many names have a stronger connection and wider range than all of them, which implies that the citation number does not directly translate into research cooperation. However, while seeking the other names on their publication (which is usually used by the project manager/coordinator of the work), names such as SCALA F., SALATINO, P., ANTHONY E. J., and ABANADES J.C. are not uncommon but are extensively interconnected and have many strong connections on the central cluster. Even though the analysis in Figure 2 initially returned authors having the largest number of citations, Figure 7 shows that citation number did not directly translate into a cooperative researcher, even though it is likely that the paper contained a strong cooperative author.

### 3.2. Comparative Analysis of Experimental Data

Table 1 presents a comprehensive analysis of relevant experimental data from the data frame. Table 1 was compiled with the purpose of presenting the main parameters used in experimental setups of fluidized bed reactors used in CaL processes. It shows that experimental setups have been unique, as there were different sizes, sorbents, heating methods, and conversion atmospheres.

#### 3.2.1. Experimental Setup

Fluidized bed reactors were classified by their flow regime, i.e., their circulating or bubbling fluidized beds. A total of 54.8% were configured as bubbling, 26.2% were configured as circulating, and 11.9% used both flow regimes, but 7.1% had no clear information about their configurations. The majority of studies used two fluidized beds for each reaction represented by Equation (1), although lab-scale studies presented a single bed configu-

ration, usually on a smaller scale. In the data frame, 52.4% of data presented a dual bed configuration where two reactors were used, and 47.6% consisted of a single reactor.

The heating method of reactors is divided into two main topics, one being electrical resistance and the other being fuel combustion. A total of 66.7% of data was on using the electrical resistance method, while 16.7% used calciner run by fuel types that included coal, hard coal, lignite, natural gas, and biomass. Tregambi et al. [41] was the only study to use a solar/infrared heating method. A total of 14.3% of studies made use of a combination of a calciner and electrical resistance to heat reactors.

Regarding the sizing of reactors, the minimum height reported from the data frame was 0.1 m, while the highest height reached 15 m, thus reaching a value of 5.5 m on average. The diameter followed a similar pattern: the values of the minimum and maximum diameters were 0.018 m and 0.75 m, respectively, thus yielding 0.22 m on average. A total of 16.7% of works used carbonators and calciners of different sizes.

Calcination and carbonation operation temperatures varies widely. Calcination temperatures were in a total range of 750–1000 °C, and carbonation temperature ranged between 550 and 622 °C.

The velocities reported on fluidized bed reactors were defined differently. Velocity is commonly referred to as the relation of superficial velocity required to start fluidization using the minimum velocity. This relation defines non-dimensional parameters, which can be used to scale up the design. In the data frame, velocities were reported as minimum fluidization (0.06–2.5 (m/s)), average velocities (0.8–2.8 (m/s)), superficial velocities (0.5–5.9 (m/s)), and times of minimum fluidization (2–7 (-)).

### 3.2.2. Sorbents

Limestone was the most common CaO-based sorbent used, as it constituted 77% of total sorbent usage. Most experimental setups used raw limestone or in pellets with the additions of cement and clay. Given the fact that it is a natural component, its composition is usually associated with its source due to different compositions. For the present data frame, it was found that 25.4% was from Germany, 14.3% was from Poland, 12.7% was not mentioned, 12.7% was from Italy and Canada, 6.3% was from China, Greece, and Spain, and 3.2% was from the UK. Other works used dolomite and lab-grade CaO, either with or without doping.

Sorbents were found to be the most widely used, and its particle size ranged from 25 to 800 µm. Furthermore, many studies use sand inside the reactor to enhance the thermal inertia of the bed. Sand was reported as inactive in the reaction and had a vastly different particle size from that of the sorbent for later sieving. Hydration was used to recover spent limestone due to losses of efficiency; a total of 37% of works made use of some method of hydration to investigate its effects or enhance capture efficiency.

### 3.2.3. Capture Efficiency

Regarding the topic of capture efficiency, it is worth mentioning that authors have used different approaches to evaluate its efficiency. Our analysis showed a set of different equations by clustering those showing slight differences. The equations are presented in chronological order below: Blamey et al. [51]  $\alpha_\infty$  is an asymptote constant;  $C_N$  is the capture capacity;  $RMM_x$  is the molar mass of species  $x$ ;  $F_x$  is the fraction of species  $x$  in the original limestone;  $k$  is the decay constant;  $N$  is the cycle number.

$$C_N = 100 \times \left[ \frac{1}{(1/(1 - \alpha_\infty)) + kN} + \alpha_\infty \right] \times \left[ \frac{RMM_{CaCO_3} F_{CaCO_3}}{RMM_{CO_2} \left( \left( \frac{F_{CaCO_3} RMM_{CaO}}{RMM_{CaCO_3}} \right) + (1 - F_{CaCO_3}) \right)} \right]$$

Blamey et al. [60]  $\alpha_\infty$  is an asymptote constant;  $k$  is the decay constant;  $N$  is the cycle number.

$$C_N = 100 \times \left[ \frac{1}{(1/(1 - \alpha_\infty)) + kN} + \alpha_\infty \right]$$

Rodríguez et al. [52]

$$E_{carb} = \frac{\text{CO}_2 \text{ reacting with CaO in the bed}}{\text{CO}_2 \text{ entering the bed in the flue gas}}$$

Coppola et al. [53]  $F_{\text{CO}_2}^{\text{in}}$  and  $F_{\text{CO}_2}^{\text{out}}$  are the inlet and outlet  $\text{CO}_2$  mass flow rates, respectively;  $m_0$  is the initial mass of sorbent in the system.

$$\text{CO}_2 \text{ Capture Capacity} = \frac{\int_0^t [F_{\text{CO}_2}^{\text{in}} - F_{\text{CO}_2}^{\text{out}}(t)] dt}{m_0}$$

Alonso et al. [61]  $F_{\text{CO}_2, \text{comb}}$  and  $F_{\text{CO}_2, \text{out}}$  are the molar flow rate of  $\text{CO}_2$  ( $\text{mol}/\text{m}^2\text{s}$ ) generated by combustion and in equilibrium, respectively.

$$E_{carb} = \frac{F_{\text{CO}_2, \text{comb}} - F_{\text{CO}_2, \text{out}}}{F_{\text{CO}_2, \text{comb}}}$$

Duelli et al. [62]  $F_{\text{Ca}}$  and  $F_{\text{CO}_2}$  are the molar flow rates of Ca and  $\text{CO}_2$  entering and leaving the carbonator, respectively;  $X_{\text{calc}}$  and  $X_{\text{carb}}$  are the contents of  $\text{CO}_2$  in Ca particles before and after the reaction, respectively.

$$E_{\text{CO}_2} = \frac{F_{\text{Ca}}(X_{\text{carb}} - X_{\text{calc}})}{F_{\text{CO}_2}}$$

Duelli et al. [63] Carbonator efficiency ( $E_{\text{carb}}$ ) is defined as the moles of  $\text{CO}_2$  captured by the solids with respect to moles introduced into the reactor.

$$E_{carb} = 1 - \frac{F_{\text{CO}_2, \text{out}}}{F_{\text{CO}_2, \text{in}}}$$

Symonds et al. [64]  $F_{\text{CO}_2}$  represents the molar flow rate of  $\text{CO}_2$  entering the carbonator ( $\text{kmol}/\text{h}$ ), and  $F_{\text{carb}}$  represents the molar flow rate of  $\text{CO}_2$  leaving the carbonator ( $\text{kmol}/\text{h}$ ).

$$E_{carb} = \frac{F_{\text{CO}_2} - F_{\text{carb}}}{F_{\text{CO}_2}}$$

Hilz et al. [44]  $F_{\text{CO}_2, \text{carb}, \text{in}}$ ,  $F_{\text{CO}_2, \text{coal}}$ ,  $F_{\text{CO}_2, 0}$ , and  $F_{\text{CO}_2, \text{calc}, \text{out}}$  refer to the  $\text{CO}_2$  fed into the carbonator, the  $\text{CO}_2$  formed by oxygen-derived fuel combustion, the  $\text{CO}_2$  from calcinated fresh limetone, and the  $\text{CO}_2$  at the calciner outlet, respectively.

$$E_{\text{total}} = \frac{F_{\text{CO}_2, \text{calc}, \text{out}}}{F_{\text{CO}_2, \text{carb}, \text{in}} + F_{\text{CO}_2, 0} + F_{\text{CO}_2, \text{coal}}}$$

Antzara et al. [18]

$$\text{CO}_2 \text{ capture, \%} = \frac{\text{moles of CO}_2, \text{in} - \text{moles of CO}_2, \text{out}}{\text{moles of CO}_2, \text{in}}$$

Blamey et al. [16]  $t$  is the carbonation time;  $X_{\text{in}}$  is the the inlet fraction of  $\text{CO}_2$ ;  $X_{\text{out}}$  is the the outlet fraction of  $\text{CO}_2$ , and  $\eta_{\text{in}}$  is the total molar flow rate of gas fed into the reactor.

$$\eta_{\text{CO}_2, \text{carb}} = \int_0^t \left( \frac{\eta_{\text{in}}(X_{\text{in}} - X_{\text{out}})}{(1 - X_{\text{out}})} \right) dt$$

Diego and Arias [15]  $E_{carb}$  is the CO<sub>2</sub> capture efficiency achieved by the carbonator;  $K_s$  is the reaction rate constant;  $\varphi$  represents the gas–solid contact factor;  $\nu_{CO_2}$  is the molar fraction of CO<sub>2</sub>, and  $\nu_{CO_2,eq}$  is the molar fraction under equilibrium conditions;  $\tau_{active}$  is the active space time;  $f_a$  is the active particle fraction;  $W_{ca}$  is the molar bed inventory of calcium in the carbonator;  $F_{CO_2,in}$  is the molar flow of CO<sub>2</sub> at the reactor inlet;  $X_{ave}$  is the average CO<sub>2</sub> capture capacity;  $t^*$  is the characteristic reaction time.

$$E_{carb} = K_s \varphi \tau_{active} (\nu_{CO_2} - \nu_{CO_2,eq})$$

$$\tau_{active} = f_a \frac{W_{ca}}{F_{CO_2,in}} X_{ave}$$

$$f_a = 1 - e^{-\frac{t^*}{W_{ca}/F_{ca}}}$$

$$t^* = \frac{X_{ave} - X_{calc}}{k_s \varphi X_{ave} (\nu_{CO_2} \nu_{CO_2,eq})}$$

Dong et al. [47]  $m_1$  and  $m_0$  are the final and initial mass of loaded samples, respectively;  $M_{CaO}$  and  $M_{CO_2}$  are the molar masses of CaO and CO<sub>2</sub>, respectively.

$$CCE(\%) = \frac{m_1 - m_0}{m_0} \frac{M_{CaO}}{M_{CO_2}} \times 100$$

Coppola et al. [33]  $W_{CO_2}^{in}$  is the mass flow rate of CO<sub>2</sub> at the carbonator inlet,  $W_{CO_2}^{out}(t)$  is the mass flow rate of CO<sub>2</sub> at its outlet,  $t$  is the carbonation time,  $MW$  is the molecular weight, and  $f_{CaCO_3}$  is the mass fraction of CaCO<sub>3</sub>.

$$X_{Ca} = \frac{\int_0^t W_{CO_2}^{in} - W_{CO_2}^{out}(t) \, dt}{m_0 f_{CaCO_3}} \frac{MW_{CaCO_3}}{MW_{CO_2}}$$

There are a huge number of unique approaches to measure carbon capture efficiency. Even though the equation could be further divided into overall, carbonator, and calciner carbon capture efficiencies, either with or without fuel usage, its main parameters were found as follows: residence time on the bed, amount of CO<sub>2</sub> introduced or generated on the bed, reaction rate, fraction of active particles, mass flow rate of reactive particles introduced onto the bed, mass flow rate of particles leaving the bed, CO<sub>2</sub> mass flow rate within the bed, and cycle number.

Such a substantial number of equations is advantageous to scale up the technology, since different approaches have been individually evidenced. However, finding matching studies is quite challenging due to the dispersion of equations, which reveal disagreement among academics. Furthermore, there is no study comparing these equations in the literature, which is suggested as further research herein.

### 3.2.4. Scientific Gaps Development

Table 1 offers a concise overview of the gaps in each article. Five primary gaps were detected in the majority of works: the use of pellets rather than raw materials, attrition and sintering, temperature effects, the CO<sub>2</sub> capture capacity mechanism, and the reactivation of sorbent and steam effects. Despite the fact that these categories are listed separately, these effects are frequently intertwined.

CO<sub>2</sub> capture capacity improvements and its effects were the most widely reported and discussed, in addition to the fact that sorbent capacities on fluidized beds tend to be lower than those reported through thermogravimetric analyses [65]. This affirmation was further ascertained by many works attributing such differences to a lack of similarities between fixed bed and fluidized bed tests [66], not to mention the distinct fluid dynamics affecting heating and reaction rates [67]. Another indirect fact worth mentioning also contributes to the affirmation that particle sizes affect fluidization due to segregation [61],

in addition to the fact that fuel quality and inert material within the bed influence sorbent inactiveness [68].

CO<sub>2</sub> capture capacity was found to be susceptible to multiple parameters, such as the presence of SO<sub>2</sub> reducing effectiveness [30,69], which could greatly affect efficiency, even in negligible amounts [30]. Soon afterwards, it was found that high temperatures and CO<sub>2</sub> concentration could affect CO<sub>2</sub> capture capacity [70]. The effect of temperature was further explored, and it was found that combustion leads to higher heat transfer, although it was also detected that it causes local high temperature spots, which, in turn, increase sintering and the decay of activities [41,50,63]. Moreover, some researchers have reported that not only high temperature causes damage in long-term carbon capture, but thermal shocks might also have an effect on particles [71]. It was also reported that this damage might be irreversible once the thermal history of the sorbent seems to affect the CO<sub>2</sub> carbon capacity [71,72].

Table 1. Data frame experimental setup parameters and conditions.

Reference (year)	Sorbent: Type/Pretreatment/Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Coppola et al. [33]	Limestone / no / Italy, Poland, Germany / 600–400 μm	Bubbling / Dual / Resistance / n.i. × 0.04 m	Calc 10%CO <sub>2</sub> -air, carb 10%CO <sub>2</sub> -N <sub>2</sub> / 950 × 650 °C / $U_{carb} = U_{calc} = 0.5$ m/s / n.i / 85–15%	(1) Materials, pre-processed in dual interconnected fluidized beds, underwent impact fragmentation following a chipping/splitting pattern as a function of an increase in impact velocity.
Coppola et al. [73]	Limestone / hydration / Italy / 600–400 μm	Bubbling / Dual / Resistance / n.i. × 0.04 m	Calc 10%CO <sub>2</sub> -Air carb 10%CO <sub>2</sub> -0-10%H <sub>2</sub> O-N <sub>2</sub> / 850 × 700–600 °C / $U_{carb} = U_{calc} = 0.5$ m/s / n.i / 0.23–0.03 g/g	(1) Optimal carbonation temperature of 650 °C is suggested for this limestone once it both satisfies CO <sub>2</sub> uptake of <i>N</i> number of carbonation stages and slight tendency to attrition/fragmentation.
Chai et al. [74]	Limestone and doped CaO / no / China / 180–125 μm	Bubbling / Single / Resistance / 0.9 × 0.032 m	CO <sub>2</sub> -CO-NO-O <sub>2</sub> -N <sub>2</sub> balance carb, 20%O <sub>2</sub> -CO <sub>2</sub> calc / 950 × 650 °C / $U_{carb} = 0.018$ m/s / n.i / n.i	(1) the Ce ((CH <sub>3</sub> CO <sub>2</sub> ) <sub>3</sub> Ce·xH <sub>2</sub> and limestone mixture) doping is more advantageous for N <sub>2</sub> formation and CO <sub>2</sub> adsorption than Mn doping on the CaO surface.
Li et al. [75]	Limestone and pellets / no / Canada / 850–600 μm	n.i / Single / Resistance / n.i × 0.0254 m	17%CO <sub>2</sub> -N <sub>2</sub> / 850 × 675 °C / n.i / n.i / 0.5–0.1 g/g	(1) Synthetic sorbents showed an uptake capacity of 0.4–0.45 g/g after 5 cycles in the reactor system, which was much greater than the uptake capacity of Cadomin limestone (0.15 g/g after 5 cycles). (2) MgO-promoted sorbent indicated a higher uptake capacity than CaZrO <sub>3</sub> -promoted sorbent due to the higher mass ratio of CaO in the sorbent. (3) SEM images indicated formation of cracks in post-cycling samples as a result of expansion and contraction of sorbent grains going through carbonation and calcination.
Hashemi et al. (2022) [76]	Limestone / no / China / 1250–150 μm	Bubbling / Single / Resistance / n.i × 0.028 m	17%CO <sub>2</sub> -N <sub>2</sub> / 920–750 × 650 °C / $3 \times U_{mf}$ / n.i / 0.5–0.1 g/g	(1) The effect of particle size on limestone calcination kinetics presents three-zone characteristics of the reaction controlling mechanism. In Zone I, particle size is <80 μm, which has little effect on consumption rate, and calcination is mainly controlled by the chemical reaction. In Zone III, particle size is >450 μm; the calcination process is mainly controlled by gas diffusion, and the effect of particle size is significant. In Zone II, particle size ranges between 80 and 450 μm and calcination is controlled by both gas diffusion and the chemical reaction.
González et al. [77]	Limestone / hydration / Canada, United Kingdom, Spain / 710–500 μm	n.i / Single / Resistance / 0.5 × 0.021 m	15%CO <sub>2</sub> -0-10%steam-N <sub>2</sub> / 900 × 700–650 °C / $8 \times U_{mf}$ / n.i / n.i	(1) Calculation results reveal that it is likely that other researchers investigating seawater and NaCl doping of limestone in CaL processes reported reductions in reactivity while overdoping their sorbents.
Haaf et al. [13]	Limestone / no / Germany / 500–25 μm	Circulating / Dual / Calciner (Fueled by Natural Gas) / 8.6 m carb and 11 m calc × 0.59 m carb and 0.4 m calc	Flue gas 9.5%CO <sub>2</sub> / 920–750 °C × 690–610 °C / $U_{carb} = 3.5$ – $2.5$ m/s / $U_{calc} = 5.5$ – $4.5$ m/s / 1 MW / 92–42%	(1) Attention should be placed upon the hydrodynamics of coupled fluidized bed systems with regard to coarse inert ash fractions. (2) Further works are required to assess the influence of solid recovered fuel during the solid phase and gaseous emission generation by the calciner.
Dong et al. [47]	Lab grade CaO / Hydration / n.i / 850–430 μm	Bubbling / Single / Resistance / 0.6 m × 0.06 m	Carb = 20%CO <sub>2</sub> -0-40%steam-N <sub>2</sub> , calc = 10–40%steam-N <sub>2</sub> / 900 × 650 °C / n.i / n.i / dry = 15–20.5%, hydrated = 15–21.7%	(1) The effect of steam during calcination was influenced by steam concentration, while carbonation conversion efficiency increased by adding 10 vol.% steam after 10 CaL cycles, a concentration of 20 and 40 vol.% led to a negative impact on sorbent reactivity due to sintering acceleration leading to smaller surface area and larger pore size.

Table 1. Cont.

Reference (year)	Sorbent: Type/Pretreatment/Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Diego and Arias [15]	Limestone / no / n.i / 130–80 μm	Circulating / Dual / Calciner (n.i) / 15 m × 0.65 m carb and 0.75 m calc	14%CO <sub>2</sub> carbonator / n.i × 670–660 °C / $U_{carb} = 5.5\text{--}2.0$ m/s and $U_{calc} = 5.9\text{--}2.6$ m/s / 1.7 MW / 100–50%	(1) Modifications to the make-up flow might be needed in a few cases to counterbalance the hydrodynamic changes occurring in flexible CaL systems to ensure high CO <sub>2</sub> capture efficiencies.
Hilz et al. [12]	Limestone / no / n.i/ n.i	Circulating / Dual / Calciner / 8.6 m carb and 11 m calc × 0.6 m carb and 0.4 m calc	Carb = 13%CO <sub>2</sub> × n.i / 900 × 700–630 °C / $U_{carb} = 2.2$ m/s and $U_{calc} = 5.5\text{--}5.0$ m/s / 1 MW / 88–70%	(1) Conservative parameters, such as specific carbonator inventory > 700 kg/m <sup>2</sup> , make-up ratio >0.15 mole Ca/mole CO <sub>2</sub> , and looping rate of 10–15 mole Ca /mole CO <sub>2</sub> were identified as the basis to scale up the process.
Coppola et al. [46]	Limestone / Hydration/ Germany / 600–400 μm	Bubbling / Dual / Resistance / 1 × 0.04 m	Calc = 70%CO <sub>2</sub> -air and carb = 15%CO <sub>2</sub> -Air, 15%CO <sub>2</sub> -10%H <sub>2</sub> O-Air / 940 × 650 °C / $U_{carb} = U_{calc} = 0.5$ m/s / n.i/ dry = 0.05–0.20 hydrated = 0.07–0.20 gCO <sub>2</sub> /g Initial sorbent	(1) Steam exerts a beneficial effect on CO <sub>2</sub> uptake, which can be even great enough to counterbalance the detrimental effect of SO <sub>2</sub> when the concentration of the latter is small. (2) With regard to sorbent mechanical properties, attrition is very limited under all operating conditions. The presence of H <sub>2</sub> O and/or SO <sub>2</sub> leads to a slight increase in sorbent fragmentation which, however, is not likely to significantly affect the operation of a CaL process.
Su et al. [78]	Limestone, cement, spent bleaching clay / no / China / 600–250 μm	Bubbling / Single / Resistance / 1.1 × 0.025 m	Carb = 15–20%CO <sub>2</sub> -N <sub>2</sub> calc = 80%CO <sub>2</sub> -N <sub>2</sub> / 920 × 650 °C / $U_{carb} = 0.05$ m/s / n.i/ 10–44 g CO <sub>2</sub> /100 g calcined sorbent	(1) The prepared pellets demonstrated lower elutriation rates, as they had greater sphericity than limestone particles. (2) Doped limestone-based pellets showed superior cyclic CO <sub>2</sub> capture capacity and attrition resistance.
Alonso et al. [42]	Limestone / no / n.i / 600–0 μm	Circulating / Dual / Resistance / 6.3 m carb, 6.1 m calc × 0.1 m	Air and coal fumes / 780 °C × n.i/ n.i/ 30 kW / n.i	(1) Neither the attrition index defined by Gwyn nor that proposed by Forsythe and Hertwig are able to account for the changes observed in power spectral density (PSD) curves sufficiently well. However, the total particle generated index and the maximum particle size generated during attrition, which require a re-construction of PSDs of elutriated and non-elutriated solids, provided a more accurate description of attrition phenomena for this methodology, in addition to distinguishing different attrition mechanisms and ranking the materials
Arias et al. [43]	n.i/ no / n.i / n.i	Circulating / Dual / calciner (Fueled by coal) / 15 m × 0.65 carb m and 0.75 calc m	Carb = 12–14%CO <sub>2</sub> and calc = 70%CO <sub>2</sub> / 950–830 × 715–620 °C / $U_{carb} = 4.5\text{--}2.5$ m/s and $U_{calc} = 5.0\text{--}3.5$ m/s / 1.7 MW / 95–40%	(1) Results confirm that it is possible to operate the calciner of a CaL system under oxygen-rich conditions due to the endothermic nature of a calcination reaction and the large flow of solids circulating within the carbonator. The axial temperature profiles measured along the calciner during these tests showed that no hot spots are found as long as there is sufficient circulation and bed inventory of solids in the calciner.
Coppola et al. [71]	Limestone / no / Germany / 600–400 μm	Bubbling / Dual / Resistance / 1 m × 0.04 m	Carb = 70%CO <sub>2</sub> -air, Calc = 15%CO <sub>2</sub> -air / 940 × 650 °C / $U_{carb} = U_{calc} = 0.5$ m/s / n.i / 0.02–0.22 g co <sub>2</sub> / g sorbent	(1) The larger thermal shocks experienced by the sorbent in single bed system tests appear to be detrimental in terms of CO <sub>2</sub> capture and attrition tendency. (2) The thermal history has non-trivial effects on sorbent fragmentation and is largely associated with the tendency of limestone sintering. (3) Sintering strengthens particle structure, which hampers secondary fragmentation on the one hand but intensifies the effects of thermal shock (primary fragmentation) on the other.

Table 1. Cont.

Reference (year)	Sorbent: Type/Pretreatment/Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Antzara et al. [18]	CaO-based, limestone / hydration / Greece / 500–355 μm	Bubbling / Single / Resistance / n.i × 0.018 m	Dry carb = 10%CO <sub>2</sub> -N <sub>2</sub> -3.2%O <sub>2</sub> -N <sub>2</sub> , hydr carb = 10%CO <sub>2</sub> -20%H <sub>2</sub> O-3.2%O <sub>2</sub> -N <sub>2</sub> dry calc = N <sub>2</sub> hydr calc = 20%H <sub>2</sub> O-N <sub>2</sub> / 920–800 × 680–650 °C / $U_{carb} = U_{calc} = 3.8\text{--}2.5$ m/s / n.i / hydr = 88–14%	(1) The addition of steam led to higher conversion rates, especially during pre-breakthrough, due to decreased CO <sub>2</sub> diffusion resistance through the formed layer of CaCO <sub>3</sub> . (2) In addition, steam significantly enhanced sorbent stability, leading to <16% deactivation after 20 consecutive cycles. (3) When the sorbent was tested in tests at slightly different temperatures for carbonation (680 °C) and calcination (750 °C), it exhibited similar carbonation conversion but had a higher deactivation. (4) Compared to a natural limestone that was used as reference material, the final capacity of CaO/CaZrO <sub>3</sub> was almost 5.6 times greater.
Hilz et al. [44]	Limestone / no / Germany / Mean 175 μm	Circulating / Dual / Calciner (Fueled by hard coal and Lignite) / 8.6 m carb 11 m calc × 0.6 m carb 0.4 m calc	n.i / 935–840 × 711–642 °C / n.i / 1 MW / 85–45%	(1) Demonstration of a successful semi-industrial pilot testing of the CaL process on a scale of 1 MWth under realistic operating conditions.
Tregambi et al. [41]	Limestone / no / Italy / 590–420 μm	n.i / Single / Solar-infrared / 0.1 m × 0.102 m	Carb = 15%CO <sub>2</sub> and calc = 70%CO <sub>2</sub> / 950–940 × 650 °C / $2 \times U_{sand,mf}$ / 3.2 kW / 0.025–0.085 g CO <sub>2</sub> / g sorbent	(1) It is inferred that local and occasional higher peak temperatures experienced by sorbent particles in solar CaL result in more extensive thermal sintering and loss of CO <sub>2</sub> capture capacity.
Hilz et al. [68]	Limestone / no / Germany / mean 175 μm	Circulating / Dual / Calciner (Fueled by hard coal and lignite) / 8.6 m carb 11 m calc × 0.6 m carb 0.4 m calc	Carbonator 11–16%CO <sub>2</sub> / 940–850 × 700–639 / $U_{carb} = 2.5\text{--}2.3$ m/s, $U_{calc} = 5.5\text{--}4.5$ m/s / 1 MW / 95–45%	(1) An accumulation of inert material shows the dependency of ash and sulphur fractions on fuel combusted in the calciner. (2) High-grade fuel reduces the inactive sorbent share in comparable operation conditions from around 20 to 8 wt.%.
Coppola et al. [72]	Limestone / hydration / Germany / 600–400 μm	Bubbling / Single / Resistance / 0.95 m × 0.040 m	Calc = 70%CO <sub>2</sub> -30%air, carb = 15%CO <sub>2</sub> -85%air hydr calc = 10%H <sub>2</sub> O-70%CO <sub>2</sub> -air hydr carb = 10%H <sub>2</sub> O-15%CO <sub>2</sub> -air / 940 × 650 / $U_{carb} = U_{calc} = 2.5\text{--}2$ m/s / n.i / dry = 0.17–0.05 and hydrated = 0.19–0.07 g CO <sub>2</sub> / g sorbent	(1) Synergistic effects were observed when steam was added, both during calcination and carbonation, thus resulting in a very pronounced increase of sorbent CO <sub>2</sub> capture capacity compared to a no-steam case. (2) A characterization of the fragmentation propensity of samples in light of morphological features assessed via microscopy showed that exposure to steam during calcination induces a more resistant external particle structure.
Coppola et al. [79]	Limestone / no / Germany / 600–400 μm	Bubbling / Dual / Resistance / 1 m × 0.04 m	100–15%CO <sub>2</sub> air / 850 × 650 °C / $U_{mf} = 0.06$ m/s / n.i / 0.05–0.20 g CO <sub>2</sub> / g sorbent	(1) CO <sub>2</sub> capture capacity results exhibited a typical decay trend with respect to the number of cycles, as expected in CaL processes. Interestingly enough, a comparison of these results with those previously obtained using the same limestone and under similar operating conditions for single bed apparatuses revealed that capture capacity values were higher than those for single bed reactors. This finding seems to emphasize a non-negligible role of sorbent thermal history in CaL CO <sub>2</sub> capture performance.

Table 1. Cont.

Reference (year)	Sorbent: Type/ Pretreatment/ Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/ Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Peng et al. [80]	CaO + Al <sub>2</sub> O <sub>3</sub> / no / China / 300– 200 μm	Bubbling / Single / Resistance / 0.892 m × 0.026 m	Carb = 10%CO <sub>2</sub> -N <sub>2</sub> , calc = 100%N <sub>2</sub> / 900 × 700 °C / U <sub>carb</sub> = U <sub>calc</sub> = 5.0–3.0 m/s / n.i / 0.45–0.82 mol CO <sub>2</sub> / mol CaO	(1) The results of ESEM prove its high sintering resistance to maintain abundant available surface area and stable textural structure for CO <sub>2</sub> capture reactions, which evidences the advantage of novel SATS method. In addition to addressing the loss-in-capacity of CaO-based CO <sub>2</sub> sorbents, attrition resistance is another important parameter worth being considered, especially in fluidized bed environments. (2) Pore size distribution and mechanical durability results show that the CaO/TiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub> sorbent exhibits high mechanical strength, long-lasting anti-attrition performance, and favorable long-life stability. Finally, a raw material analysis shows that the CaO/TiO <sub>2</sub> -Al <sub>2</sub> O <sub>3</sub> sorbent has a competitive advantage for further continuous long-term large-scale industrial operations.
Ridha et al. [81]	Cadomin and Lime- stone Pellet / no / Canada, Spain / 600–212 μm	Mixed / Dual / Resistance and calciner (natural gas carbonator and calcined biomass wood pellets) / 5.1 m calc, 3 m carb × 0.1 m	80%CO <sub>2</sub> / 900 × 650 °C / U <sub>carb</sub> = 0.8–0.5 m/s, U <sub>calc</sub> = 2.6–2.0 m/s / 0.1 mW / 90–80%	(1) Batch and continuous injection of pellets into the system revealed that the injection method exerted an insignificant effect on the attrition of pellets. The similarity of particle size distribution patterns of pellets is indicative of similar attrition tendencies, and it appears to be a characteristic of these pellets, regardless of limestone type and injection method. (2) It was found that the pelletization of CaO-based sorbents using limestone and 10% cement results in a marginal improvement in the mechanical strength of resultant pellets. Therefore, the pelletization of sorbent for CaL CO <sub>2</sub> capture is considered inadequate for sorbent attrition reduction.
Symonds et al. [64]	Cadomin and Lime- stone Pellet / no / Canada, Spain / 600–250 μm	Mixed / Dual / Resistance and Calciner (natural gas carbonator and biomass pellets calciner) / 5.1 m calc, 3 m carb × 0.1 m	80%CO <sub>2</sub> / U <sub>carb</sub> = 0.8–0.5 m/s, U <sub>calc</sub> = 2.6–2.0 m/s / 0.1 mW / 100–82%	(1) In accordance with published works on CO <sub>2</sub> capture performance of synthetic/modified CaO-based sorbents at bench-scale, this work aims to highlight the fact that such sorbents could have had a more significant and enhanced performance when tested in a pilot-scale system. This suggests that the assessment of such relatively expensive sorbents should be performed under pilot-scale testing conditions to evaluate their performance with respect to their production costs.
Blamey et al. [16]	Limestone and cement pellets / hydration / n.i / 710–500 μm	Bubbling / Single / Resistance / n.i × 0.021 m	15%CO <sub>2</sub> -N <sub>2</sub> and 10%H <sub>2</sub> O13.5%CO <sub>2</sub> N <sub>2</sub> / 900 × 700–600 °C / U <sub>carb</sub> = 7.2–5.8 m/s, U <sub>calc</sub> = 9.8–7.9 m/s / n.i / 0.32–0.08 g CO <sub>2</sub> / g calcined sorbent	(1) Bubbling bed reactor tests were carried out using up to 20 CO <sub>2</sub> capture cycles on limestone-based pellets produced using calcium aluminate as the binder. These tests show that pellets exhibit a similar or slightly more enhanced behavior in terms of attrition resistance to its parent limestone, in addition to showing superior CO <sub>2</sub> capture performance altogether.

Table 1. Cont.

Reference (year)	Sorbent: Type/Pretreatment/Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Coppola et al. [82]	Limestone / hydration / Germany / 600–400 μm	Bubbling / Single / Resistance / n.i × 0.04 m	Calc = 70%CO <sub>2</sub> -air, carb = 15%CO <sub>2</sub> -Air, Hydration = 50%H <sub>2</sub> O/N <sub>2</sub> / 940 × 650 °C / $U_{carb} = U_{calc} = 2.5\text{--}2.0\text{ m/s}$ / n.i / spent = 0.10–0.48 and hydrated = 0.11–0.51 mol/mol	(1) The effectiveness of sorbent hydration using steam as a means to reactivate CO <sub>2</sub> uptake potential of limestone for CaL applications has been demonstrated. Steam hydration followed by dehydration of reactivated sorbent in a hot fluidized bed reactor leads to increased porosity and, hence, an improved rate and length of CO <sub>2</sub> uptake. (2) Furthermore, attrition and fragmentation propensity of reactivated sorbent is increased. (3) For the aforementioned limestone, optimal trade-off between sorbent reactivity/uptake and mechanical strength is achieved after 60 min of hydration of spent sorbent, but it is expected that such a result cannot be generalized, since it is critically dependent upon sorbent texture. A comparison between water and steam hydration of spent sorbents as means of reactivation indicated that steam hydration is more favorable. (4) Albeit liquid water hydration gives rise to greater water uptake, prolonged soaking of liquid water makes the reactivated sorbent more susceptible to attrition and fragmentation.
Duelli et al. [63]	Limestone / hydration / Germany / 375 μm	Mixed / Dual / Resistance / 12.4–0.5 × 0.15–0.071 m	Calc = 0–75%CO <sub>2</sub> –0–35%H <sub>2</sub> O–N <sub>2</sub> , carb = 10–16CO <sub>2</sub> –10H <sub>2</sub> O–N <sub>2</sub> / 930–880 × 680–600 / $U_{carb} = 5.5\text{--}4.5\text{ m/s}$ / $U_{calc} = 2.0\text{--}0.3\text{ m/s}$ / 10 kW / dry = 60–20% hydr = 95–40%	(1) Combustion may lead to faster calcination due to better heat transfer; however, local high temperatures may cause pronounced sintering and consequent further decay of activity as recorded herein, and there might be sorbent hardening.
Duelli et al. [62]	Limestone / no / Germany / n.i	Bubbling / Dual / Resistance / 12.4 m × 0.114 m	Calc = 0–65%CO <sub>2</sub> -N <sub>2</sub> , carb = 14%CO <sub>2</sub> -N <sub>2</sub> / 920–905 × 630 / n.i / 10 kW / 18–9% mol CaCO <sub>3</sub> /mol Ca	(1) Full sorbent regeneration could not be achieved primarily due to the bubbling fluidization regime of the bed and the quality of heat provided by electric heaters through the walls to the bed. These limitations impose solid residence times of up to 8 min for a calcination of solids instead of seconds, as it is in TGA investigations as well as industrial applications such as clinker production. This is not the case for process scale-up where these limitations are not found due to a fast fluidization regime and combustion atmosphere; thus, the data presented herein are suggested to be treated qualitatively, not quantitatively.
Coppola et al. [83]	Limestone / hydration / Italy / 600–400 μm	Bubbling / Single / Resistance / n.i × 0.04 m	70%CO <sub>2</sub> -Air / 940 × 650 °C / $U_{mf} = 0.7\text{ m/s}$ / n.i / hydr = 0.03–0.35 gg <sup>-1</sup> , dry = 0.03 gg <sup>-1</sup>	(1) Optimal hydration time depends on pore size.
Alonso et al. [61]	Limestone / no / Spain, Germany / 348–87 μm	Circulating / Dual / Calciner (biomass pellets) / 12 m × 0.4 m	n.i / 950–800 × 720–630 °C / $U_{carb} = U_{calc} = 2.8\text{--}0.8\text{ m/s}$ / 300 kW / 85–5%	(1) Large discrepancies were detected in experiments conducted with large particles, probably due to segregation effects of denser and coarser particles in the reactor bed inventory.

Table 1. Cont.

Reference (year)	Sorbent: Type/Pretreatment/Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Coppola et al. [67]	Dolomite, Limestone / no / Italy, Germany, Greece, Poland / 600–400 μm	Bubbling / Single / Resistance / 0.95 m × 0.04 m	Calc = 70%CO <sub>2</sub> N <sub>2</sub> , carb = 15%CO <sub>2</sub> N <sub>2</sub> / 940 × 650 °C / $U_{carb} = U_{calc} = 0.7\text{--}0.6$ m/s / n.i / 0.02–0.14 g CO <sub>2</sub> / g Ca	(1) TG and FB devices are characterized by quite different fluid dynamic and mass transfer conditions, which result in different sample heating and reaction rates. However, as far as CO <sub>2</sub> capture capacity is concerned, the results suggest that the type of reactor has a lower influence than the calcination environment on sorbent performance. Factors enhancing sintering (e.g., high temperature and CO <sub>2</sub> concentration) severely impact the sorbent CO <sub>2</sub> capture capacity. On the other hand, factors slowing down sintering (e.g., the presence of MgO, as in dolomite) improve sorbent performance.
Coppola et al. [70]	Dolomite, Limestone / no / Italy, Germany, Greece, Poland / 600–400 μm	Bubbling / Single / Resistance / 0.95 m × 0.04 m	Calc = 70%CO <sub>2</sub> 30%N <sub>2</sub> , Carb = 15%CO <sub>2</sub> 85%N <sub>2</sub> / 940 × 650 °C / $U_{carb} = U_{calc} = 0.7\text{--}0.6$ m/s / n.i / 80–18% mol/mol	(1) The CO <sub>2</sub> capture capacity of the dolomite was always greater than that of limestone and remained relatively high along cycles, despite the lower calcium content of the sorbent.
Coppola et al. [53]	Limestone / no / Italy, Germany, Greece, Poland / 500–30 μm	Bubbling / Single / Resistance / 0.95 m × 0.04 m	Calc = 70%CO <sub>2</sub> 30%N <sub>2</sub> , carb = 15%CO <sub>2</sub> 85%N <sub>2</sub> / 940 × 650 °C / $U_{carb} = U_{calc} = 0.7\text{--}0.6$ m/s / n.i / capture capacity = 0.02–0.21 g CO <sub>2</sub> / g CaCO <sub>3</sub>	(1) The measured fine elutriation rate was only moderately large during the first cycle, but it reduced along the cycles, since a combined chemical thermal treatment significantly hardened the particle structure. (2) A high SO <sub>2</sub> concentration had a detrimental effect on the CO <sub>2</sub> capture capacity of all limestones, while a low SO <sub>2</sub> concentration had a more limited effect.
Coppola et al. [30]	Limestone / no / Italy / 600–400 μm	Bubbling / Single / Resistance / 1 m × 0.04 m	Calc = 20CO <sub>2</sub> N <sub>2</sub> , Carb = 16CO <sub>2</sub> N <sub>2</sub> / 850 × 700 °C / $U_{carb} = U_{calc} = 0.7\text{--}0.6$ m/s / 4.4 kW / 60–10%	(1) Results showed that the presence of SO <sub>2</sub> in the flue gas significantly decreased the sorbent CO <sub>2</sub> capacity, quite possibly due to the formation of an impervious CaSO <sub>4</sub> layer at the periphery of particles. (2) Even a small quantity of SO <sub>2</sub> is sufficient to significantly hinder the sorbent CO <sub>2</sub> capture capacity. (3) An analysis of particle size distribution of bed material over repeated calcination/carbonation cycles indicated that particle fragmentation was very limited in all investigated conditions. (4) Results obtained using 1800 ppm SO <sub>2</sub> showed divergences from other conditions with regard to: cumulative particle size distribution of fragments, cumulative elutriated material over five repeated cycles, and degree of SO <sub>2</sub> uptake over repeated cycles.
Acharya et al. [69]	Limestone / n.i / n.i / 325–135 μm	Mixed / Dual / Resistance / n.i × 0.101 m	Calc = Steam+Air Carb = 22–25%CO <sub>2</sub> -N <sub>2</sub> / 900 × 600–550 °C / n.i / 100–10% (air) and 100–25% (steam)	(1) The CO <sub>2</sub> absorbing capacity of CaO decreases progressively as it goes through alternating cycles of carbonation and calcination. Such a reduction is due to several factors, including sintering and pore pluggage. (2) The calcination of CaCO <sub>3</sub> in the presence of different media shows that the degree of calcination of CaCO <sub>3</sub> in the presence of H <sub>2</sub> O is similar to that of N <sub>2</sub> . (3) Kinetic rates of calcination are much higher when measured using H <sub>2</sub> O and N <sub>2</sub> than when measured using CO <sub>2</sub> . (4) Sintering leading to sorbent agglomeration cannot be avoided during calcination at higher temperatures, even while using steam.

Table 1. Cont.

Reference (year)	Sorbent: Type/Pretreatment/Origin/Particle Size	Fluidized Bed: Type/Dual or Single/Heating Method/Height × Diameter	Process: Calciner × Carbonator Atmosphere/Calciner × Carbonator Temperature/Investigated Velocity/Wattage/Capture Efficiency	Gaps
Symonds et al. [66]	Limestone / hydration / Canada, Poland / 612–512 μm	Circulating / Single / Resistance + Calciner (Pure fiber hardwood blend) / 5 m × 0.1 m	Calc = 1%CO <sub>2</sub> 7.5%O <sub>2</sub> 35%CO <sub>2</sub> or 1%CO <sub>2</sub> 6%O <sub>2</sub> 35%CO <sub>2</sub> and Carb = 8%CO <sub>2</sub> 19.3%O <sub>2</sub> 72.7%N <sub>2</sub> or 8%CO <sub>2</sub> 15.7%O <sub>2</sub> 17%H <sub>2</sub> O 59.3%N <sub>2</sub> / 920–860 × 690–610 / n.i / 49–11%	(1) Results suggest that CO <sub>2</sub> capture cycles in TGA experiments should encompass as many factors present during FBC operation as possible (i.e., higher calcination temperatures/CO <sub>2</sub> concentrations, and the presence of steam and other gases such as SO <sub>2</sub> ) so as to avoid rather misleading results and conclusions regarding sorbent performance.
Rodríguez et al. [52]	Limestone / no / n.i / <350 μm	Circulating / Dual / Resistance / calc = 6 m, carb = 6.5 m × 0.1 m	Calc = 5–3%O <sub>2</sub> 7–22%CO <sub>2</sub> , Carb = 15–2%CO <sub>2</sub> 17%O <sub>2</sub> / 850–800 × 722–568 °C / n.i / 30 kW / 92–30%	(1) When there are enough active CaO particles inside the reactor, the low-carrying capacity of highly cycled CaO particles can be counterbalanced by sufficiently large inventories of solids in the carbonator, which can increase the carbonation conversion achieved by CaO to values close to the maximum capture capacity of the material by increasing the average residence time of particles in the reactor.
Blamey et al. [60]	Limestone / hydration / Canada / 710–500 μm	Bubbling / Single / Resistance / 0.33 m × 0.026 m	15%CO <sub>2</sub> N <sub>2</sub> / 950–850 × 700 °C / U <sub>mf</sub> = 8 / n.i / 70–10% fresh, 35–0% hydr after 13 cycles	(1) Severe temperatures in calcination cause particles to become friable and increase elutriation process. (2) Results indicated that reactivation would be unsuitable for a bubbling fluidized bed CO <sub>2</sub> capture process, though it may be suitable for a moving or fixed bed reactor.
Blamey et al. [84]	Limestone / hydration / United Kingdom, Canada, Spain / 710–500 μm	Bubbling / Single / Resistance / 0.33 m × 0.026 m	15%CO <sub>2</sub> N <sub>2</sub> / 1000–840 × 700 °C / U <sub>mf</sub> = 7 / n.i / 78–2 g/100 g and 58–18 g/100 g hydrated	(1) Significant expansion and break-up of particles was observed in the hydration for particles cycled at higher temperatures, and it is proposed that this is due to a significant densification of particles.
Charitos et al. [50]	Limestone / no / Germany / n.i	Bubbling / Dual / Resistance and calciner (Natural gas) / 12.4 m × 0.071 m calc 0.114 m carb	15%CO <sub>2</sub> air / n.i × 700–630 °C / n.i / 10 kW / 97–90%	(1) The degree of sorbent particle sintering increased along carbonation–calcination cycles and resulted in smaller values of average CO <sub>2</sub> capture capacity, reduction in small pore volume (<200 nm), and a decrease in particle surface area.
Hughes et al. [65]	Limestone / no / Poland, Canada / n.i	Mixed / Dual / Resistance and Calciner (Wood pellets) / 5 m × 0.1 m	Calc = 46%N <sub>2</sub> O <sub>2</sub> Carb = 8%CO <sub>2</sub> Air or 8%CO <sub>2</sub> 17%H <sub>2</sub> OAir / 910–860 × 600 °C / n.i / 75 kW / n.i	(1) Sorbent capacity was significantly lower than that expected based on previous thermogravimetric analyses. It is believed to be at least partially captured due to the formation of a thin, low-porosity shell around the sorbent enhanced by the deposition of ash from the solid fuel under oxygen-fired conditions.
Lu et al. [49]	Limestone / no / Canada / 800–400 μm	Circulating / Dual / Resistance and Calciner (biomass or coal) / 5 m × 0.1 m	15%CO <sub>2</sub> air / 950–850 × 720–580 °C / U <sub>carb</sub> = U <sub>calc</sub> = 0.8–0.4 m/s / 75 kW / 85–70%	(1) An examination of sorbent surface characteristics suggests that a number of complicated processes are occurring on the particle surface as a consequence of the number of reaction cycles. Issues of sorbent loss through attrition, impact of sulphation, and process optimization require further investigation.

n.i. means nothing informed or it was not possible to find the information.

A hindrance to CaL technology is a reduction in CO<sub>2</sub> carbon capacity with a greater number of cycles due to sintering and pore plugging [69]. Sintering processes are directly linked to attrition and sorbent loss during fluidized bed reactions [49]. Notwithstanding the fact that the effects of elutriation are greater with a large number of cycles, high attrition regimes, as those found while using bubbling fluidized beds, have been reported to worsen the effect [60]. Multiple works have attempted to mitigate the effect of elutriation and sintering in order to improve CO<sub>2</sub> capture capacity. Increasing the presence of MgO using dolomite has proved to achieve higher CO<sub>2</sub> capture capacity [70]. Furthermore, counterbalancing the loss of efficiency with a larger number of particles [52], as well as the pelletization of limestone and CaO [16], have showed a few advantageous effects. Pellets are an alternative in CaL research due to achieving better performance than raw limestone [16,44]. Although some works have pointed out that the cost of pellets could be reduced on a larger scale [64], others express disagreement with [81].

The hydration of new and spent limestone is deemed as a solution to the decay effect after a large number of carbon capture cycles [46,72]. The most commonly reported reactivation method is water soaking or using steam, and the latter is reported as the most efficient [82]. Studies on salt water doping are being conducted, but it could be harmful to CO<sub>2</sub> capture capacity if overdoped [77]. Even though steam is beneficial to CO<sub>2</sub> uptake [46,72], there seems to be a lack of understanding about its effect, as some authors reported that hydration increases particle breakage at higher temperatures [84]; nevertheless, it depends on steam concentration [47].

Despite the fact that all publications on the data frame are experimental and include an investigation of the influence of modifying parameters on the CaL process, no clear mention of DoE methodologies or optimization was found.

### 3.3. Data Analysis Limitations

Although two different databases were used (Scopus and Web of Science) to search keywords used in articles about the present topic, a search on the Web of Science found only two new articles, which had a negligible impact on general results.

It was found that many authors had a problem with name spelling, which had to be handled manually, i.e., with different spelling, missing surnames, and name order. Both databases have different patterns for exporting data, and organizing both into a single data frame was quite challenging, as such a complication represents a barrier to automating data processing. Data standardization might have a beneficial impact on works such as the one herein, thus making the analysis effortless and its results more accurate.

A few changes could have impacted this analysis, i.e., standard name and position of columns storing data, file type, standard encoding, and a rigorous process for checking authors' critical information. Changes on the databases lead to an unfortunate need to include future information, which might be required; however, such changes should be informed beforehand, as it takes time for a meta-analysis algorithm to adapt to upcoming changes.

Despite the fact that a considerable effort was made to cover all literature works regarding the present topic, a few articles could not be found on the initial search. It seems as though such papers have all been published after 2008, and their keyword use differs from those reported on the present data frame. Furthermore, any paper included or excluded in searches performed after this research is published might lead to repeatability issues in papers making use of novel meta-analyses. Science databases should be perfectly stable, or as stable as possible at the least.

## 4. Conclusions

For significant advancement, there must be a clear and precise technology development direction. Given the urgency of climate change, the development of CO<sub>2</sub> capture technology should be as precise as possible in order to scale up facilities to a meaningful level. The identification of technological restrictions and barriers could aid in the production of high-quality and relevant papers. Given the above, studies collecting thorough and

valuable data on challenges may assist researchers in determining the most effective path to take. This study sought to assess the use of fluidized bed reactors using CaL technology from data available on the Scopus and Web of Science databases, and its eligibility criteria were applied to set up our database to comprise the most relevant articles on the topic under evaluation.

The data frame analysis has led to the following findings. A successful database was compiled, which showed a rising trend of paper production, even though the study was affected by the COVID-19 pandemic. Novel experimental setups were found in three periods of interest (2008, 2010–2012, and 2018), such as the SO<sub>2</sub> elutriation influence on the CaL process. The EU leads the research rankings on using fluidized bed reactors through CaL carbon capture technology. Canada has the highest ratio of publication/citation, possibly due to the nation's large investments made on carbon capture technologies. Keyword choice appears to affect the number of paper citations. A data frame analysis revealed that two to three keywords addressing the main subject are enough to delimit a search topic, and all remaining keywords should be used to highlight addressing gaps and barriers. Data standardization should be beneficial for carrying out data meta-analyses. Names of the same authors presented misspelling, missing names, or were identified differently, which could have led to data evaluation errors. Furthermore, science databases should be as stable as possible.

Recent research makes use of limestone, which varies according to its origin, such as CO<sub>2</sub> sorbent, but there are few exceptions. The experimental setup trend is to use two fluidized bed reactors individually for carbonators and calciners using different fluidizing regimes. Industrial-size reactors seem to represent the current trend; thus, they ought to be used to scale up the technology effectively. Heating methods vary based on reactor size, being electrically heated for smaller sizes and using a calciner for larger sizes. Recent literature gaps reveal that there should be further investigations into higher CO<sub>2</sub> capture efficiency and the use of steam generators, SO<sub>2</sub>, temperature, or a combination of the effect on the processes of elutriation, sintering, and performance.

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**Data Availability Statement:** Publicly available datasets were analyzed in this study. This data can be found here: <https://www.scopus.com> and <https://www.webofscience.com>.

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