

Article

Carbon Dioxide Uptake in the Roadmap 2050 of the Spanish Cement Industry

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Abstract: The European Green Deal and its endeavors will make rapid and far-reaching decisions with major implications for the European cement industry in the short- and longer-term. Accordingly, new measures should be dealt with quickly and effectively to minimize the adverse impact on global warming and global climate change by this sector. The aim of this study is to show and assess the measures to be undertaken to reach carbon neutrality by the Spanish cement industry by 2050. They may be categorized into three broad types based on the main materials: clinker, cement, and concrete. The cement sector must implement breakthrough initiatives, inventions, and technologies regarding the clinker and cement production processes. Furthermore, carbon dioxide uptake by cement-based materials must be considered to achieve the carbon neutrality objective. Accordingly, two methodologies named simplified and advanced, consistent with Guidelines for National Greenhouse Gas Inventories elaborated by the Intergovernmental Panel on Climate Change (IPCC), were selected to model the carbon offsetting by mortars and concretes. Finally, the existing climate change mitigation technologies available in Spain are insufficient to reach the net zero carbon footprint. Therefore, breakthrough technologies such as novel and efficient carbon dioxide capture, utilization, and storage (CCUS) technologies should be implemented by the Spanish cement industry to achieve zero carbon dioxide emissions in 2050.

Keywords: cement; energy-intensive industry; measures; carbon dioxide uptake; Roadmap 2050; policy

1. Introduction

1.1. The Cement Industry in the European Green Deal Context

Throughout history, Europe has been a forerunner in industrial innovation. Currently, The European Union (EU) is committed to the Paris Agreement and its greenhouse gas (GHG) emissions reduction targets [1]. Therefore, it calls for measures to be taken considering the latest available technical literature including the Intergovernmental Panel on Climate Change (IPCC) reports [2]. Accordingly, the new Industrial Strategy for Europe published in 2020 [3] highlight the main challenge that EU industries will be facing, i.e., carbon neutrality. Then, different measures to support the industry, in general, and the energy-intensive industries, in particular, are currently being studied by the European Commission. As the energy-intensive industries are essential to Europe's economy, their modernization and decarbonization are strategic issues [3,4].

Some action plans for circular economy, critical raw materials and so on, are being developed within the European Green Deal context [4]. The European cement industry has joined the plan of the European Union and the European Green Deal objectives [5]. Accordingly, carbon neutrality will be achieved through the entire cement and concrete value chain by 2050.

The European Green Deal [4] and its initiatives will make a significant impact in the European cement industry in the short and medium term. With regard to the circular economy action plan [6], reduction of natural resources usage, minimization of wastes generation and increasing circularity in the EU is promoted. The cement sector will play an important role in this topic due to the well-known use of industrial wastes to produce Portland clinker and cement [7].

For the first time, the European Commission, by means of the Circular Economy Action Plan, includes carbon removals through long term storage in products such as mineralization in building materials [5]. This is great news for the cement and concrete sectors. In addition, it is mentioned that a regulatory framework for the carbon removals certification should be ready in 2023.

Carbon neutrality transition from 2020 to 2050 demands efforts from all stakeholders. This challenge will require innovative adaptation, not only in the cement sector, but also in the concrete and construction sectors. It is intended to reduce the carbon emissions at each stage of the Portland cement and concrete lifecycle, by using appropriate technical measures, to act consistently with the European Green Deal policy [4] to reach zero emissions in 2050. For instance, in the clinker and cement production stages, the cement industry will require availability to new raw materials (decarbonated and wastes) and renewable energy. In addition, a complementary regulatory framework is necessary to enhance this transition process. Such transformation will need EU funding and new ways of financing including but not limited to taxonomy [8].

New measures must be undertaken by the Spanish cement industry to minimize its impact on the climatic change. It is necessary to assess the need to implement new and innovative measures in the cement industry to deliver carbon neutrality in 2050. Another important point is to get a reliable estimation of the carbon dioxide uptake by cement-based materials. Then, this paper shows the Spanish cement industry path towards carbon neutrality in 2050 throughout the value chain.

1.2. Carbon Dioxide Uptake by Cement-Based Materials

As climate change is one of the major societal challenges [4], the cement sector will implement some initiatives to cut carbon dioxide emissions and enforce mitigation strategies. Carbon dioxide uptake by mortars and concretes carbonation has excited much interest in recent years. This physico-chemical process can be considered as a mitigation measure because carbonation can be promoted in recycled and demolished concrete, as well as for some concrete structures [9]. At the dawn of the 21st century, Gajda and Miller [10] estimated that concretes are able to absorb about 7.6% of the carbon dioxide released during the calcination process in the clinker production. They assumed complete hydration (hydration degree = 100%) and carbonation (degree of carbonation, DoC = 100%), but only $\text{Ca}(\text{OH})_2$ carbonates. Therefore, the calcium oxide available for carbonation represents only 38% of the total CaO in the clinker. In addition, only the service-life period was considered in the calculations. Subsequently, Pade and Guimaraes [11] proposed that 100% of the CaO present in the AF_m , AF_t , and $\text{Ca}(\text{OH})_2$ and 50% of the CaO present in the C-S-H can be carbonated. Accordingly, 75% of the total CaO in the clinker is available to be carbonated. Moreover, they assumed that the Portland cement is fully hydrated, and the carbonation degree is 100%; besides, the carbon dioxide diffusion coefficient and its concentration are constant for all the situations. However, it is well-known that in most of the cases the hydration degree is lower than 100% (about 70%–80% depending on the water/cement ratio, curing conditions, and so on [12]). Then, their results could be overestimated. Furthermore, they suggested that all the calcination carbon dioxide can be fully uptake by carbonation when service life and end-of-use periods are considered. For instance, they estimated a percentage of 57% of carbon dioxide uptake with regard to the calcination emissions taken into account service-life (70 years) and end-of-use (30 years) periods (24% and 33%, respectively) for Denmark.

Recently, the “re-carbonation project” sponsored by The Portland Cement Association (PCA), The European Cement Association (CEMBUREU), The Cement Sustainability Initiative (CSI), The Global Cement and Concrete Association. (GCCA), and Cementsa (HeidelbergCement Group) [13] has proposed a simplified methodology for the service life and the end-of-life and secondary usage stages. This methodology has been applied to the Spanish mortars and concretes [14].

Summing up, Table 1 compares the carbon dioxide absorption obtained in several studies published from 2000 to 2010 (initial studies). In the first column on the right is indicated the information presented in the table: authors, year of publication, country of the researchers and the methodology to perform the estimations, i.e., life cycle or service life approach. The second column shows if the carbon dioxide is related to the calcination process or to the clinker production (calcination plus fuel combustion). The first Spanish study showed the lowest carbon dioxide uptake [15] due to the low surface/volume ($S/V = 3$) and degree of carbonation measured at one year of exposure ($DoC = 15\text{--}20\%$) considered.

Table 1. Carbon dioxide uptake estimations presented in several papers published in the first decade of the 21st century, i.e., initial studies (2000–2010), %.

Authors	CO ₂ Uptake ¹	Gajda and Miller [10]	Jacobsen and Jahren [16]	Pade and Guimaraes ² [11]			Galán et al. ² [15]
Year		2000–2001	2002	2003			2009
Country		USA	Norway	DK	NO	SU	IS
							Spain
Life cycle	calcination clinker		14–19 8–11	57 34	33 19	33 19	34 20
Service life (50–100 y)	calcination clinker	7.5 4.4		24 14	16 9	18 11	28 17
							3 2

¹ CO₂ uptake (% with regard to the total emissions or calcination process); DK: Denmark, NO: Norway, SE: Sweden, IS: Iceland; ² Surface/Volume = 3 and degree of carbonation, DoC = 15–20%.

Table 2 shows some results obtained in several studies published from 2010 to 2020 (recent studies) on carbon dioxide uptake [12,17–22]. In this table, the Spanish study has been updated considering a degree of carbonation measured at four years of exposure, $DoC = 62.5\%$. In general, it can be observed that carbon dioxide uptake, considering the whole life cycle of the product, goes from 14% [16] to 54% [11]. Accordingly, a wide range of percentages is available. These studies, in fact, suggest that it is necessary to perform local studies to achieve reliable data on carbon dioxide uptake from all around the world.

Table 2. Carbon dioxide uptake estimations presented in several papers published during the second decade of the 21st century, i.e., recent studies (2011–2020), %.

Authors	CO ₂ Uptake ¹	Nygaard and Leemann [17]	Anderson et al. [18]	Yang et al. [19]	Fitzpatrick et al. [20]	Xi et al. [21]	Vermeulen ² [22]	Andrade and Sanjuán ³ [12]
Year		2012	2013	2014	2015	2016	2017	2018
Country		Switzerland	Sweden	Korea	Ireland	Global study	Netherlands	Spain
Life cycle	calcination clinker		27 17	29–34 18–21		43 28	37 23	
Service life (40–100 y)	calcination clinker	15–16 9–10	23 14	10 6	15 10		30 19	11 7

¹ CO₂ uptake (% with regard to the total emissions or calcination process); ² Includes CO₂ uptake in slag;

³ Surface/Volume = 3, Degree of carbonation, DoC = 62.5%.

Currently, the net carbon dioxide emissions (amount emitted in the calcination process in the clinker production minus the carbon dioxide uptake occurred by the mortar and concrete carbonation) are not included in the IPCC’s Guidelines for National Greenhouse Gas Inventories [23,24]. Therefore, the net emissions should be implemented in the climatic models reported in the upcoming IPCC’s Assessment Report, and subsequently, in the IPCC’s Guidelines for National Greenhouse Gas Inventories.

2. Methodology

The European Green Deal [2] and its endeavors will make rapid and far-reaching decisions with major implications for the European cement industry in the short- and longer-term. Accordingly, new measures should be addressed effectively and rapidly to reduce, as much as possible, the adverse impact on the global warming by the cement sector, i.e., minimize its contribution to the global climate change. The aim of this study is to show and assess the measures to be undertaken to reach carbon neutrality by the Spanish cement industry by 2050 and focus on carbonation of cement-based materials as part of the Roadmap 2050 of the Spanish Cement Industry.

Measures to be undertaken for having a net zero carbon footprint may be categorized into three broad types based on the main materials: clinker, cement, and concrete. Apparently, the cement sector should embrace breakthrough initiatives, inventions, and technologies regarding the clinker production process, such as carbon dioxide capture, utilization, and storage (CCUS) technologies. Furthermore, high contents of new cement constituents should be utilized. In addition, carbon dioxide uptake by cement-based materials must be considered to achieve the carbon neutrality objective. Consequently, two methodologies named simplified and advanced, consistent with Guidelines for National Greenhouse Gas Inventories elaborated and published by the Intergovernmental Panel on Climate Change (IPCC), were selected to model the carbon offsetting by mortars and concretes in Spain.

The simplified calculation for estimating the annual carbon dioxide uptake (ACDU) is based on Equations (1) and (2), and it has been used to the 2005–2015 Spanish cement production [14]. The parameters for α and β were 0.20 and 0.03, respectively.

$$\text{ACDU (service life)} = \alpha \times \text{IPCC reported emissions due to the calcination process} \quad (1)$$

$$\text{ACDU (end-of-life)} = \beta \times \text{IPCC reported emissions due to the calcination process} \quad (2)$$

3. Results and Discussion

Cement carbon dioxide direct emissions decreased from 815 to 679 kg CO₂/t cement between 1990 and 2018 in Spain, while the sum of direct and indirect emissions was 729 kg CO₂/t cement in 2018. Only a reduction of 17% was made in about 30 years.

Direct carbon dioxide emissions come from sources controlled by the reporting cement producers, such as emissions from kiln fuels related to clinker production, combustion of organic carbon from raw materials, and calcination of carbonates present in the raw material. On the other hand, indirect carbon dioxide emissions refer to activities controlled by third parties, such as external production of electricity consumed by cement factories and transport of materials by other entities (raw materials, fuels, cement, and clinker).

In Europe, direct emissions decreased from 783 to 667 kg CO₂/t cement (15%) over the same period. On the other hand, the global GHG emissions in the European Union decreased by 23% and the economy grew by 61% in that time period, whereas global GHG emissions were reduced by 2% from 2017 to 2018. In addition, The European Union will legislate to cut down global GHG emissions by at least 40% compared to 1990 GHG emissions by 2030 [25]. Consequently, it is necessary to establish an adequate policy framework to promote new procedures and technologies that will allow to the cement industry a great reduction in carbon dioxide emissions [26]. Such a reduction of 40% of CO₂ emissions by 2030 and 100% by 2050, will be performed by the cement sector along the cement and concrete value chain. Then, the initial hypothesis to undertake the analysis is based on the 2018 scenario and the values shown in Table 3 and considering that the Spanish cement production is still growing from 0.0156 Gt/year (2018) to 0.0255 Gt/year by 2050. On the other hand, the world global cement production is expected to grow from 4.65 Gt/year (2016) to 4.85 Gt/year in 2050.

Table 3. Values of the key parameters based on the 2018 scenario (Initial hypothesis).

Material	Parameter	Units	Value
Clinker	Process emissions	kg CO ₂ /t clinker	525
Clinker	Fuel emissions	kg CO ₂ /t clinker	288
Clinker	Total net emissions ¹	kg CO ₂ /t clinker	818
Clinker	Total gross emissions (including total electricity on clinker basis)	kg CO ₂ /t clinker	842
Cement	Clinker factor	kg clinker/t cement	828
Cement	Emission factor	kg CO ₂ /t cement	679
Cement	Emission factor (including electricity and transport)	kg CO ₂ /t cement	729
Concrete	Carbon dioxide index	kg cement/m ³ /MPa	8
Concrete	Cement content	kg cement/m ³ concrete	300
Concrete	Emission factor	kg CO ₂ /m ³ concrete	200

¹ The Taxonomy Technical Report gives a threshold level for these parameters [8,27].

3.1. Clinker Measures

Figure 1 shows the carbon dioxide emission intensity in the Spanish cement industry for white and gray clinker from 2005 to 2018. As expected, the white clinker has a higher emission intensity factor than gray clinker, both for fuel combustion (white: 0.48–0.56; gray: 0.28–0.33) and for the calcination process (white: 0.54–0.52; gray: 0.53–0.52).

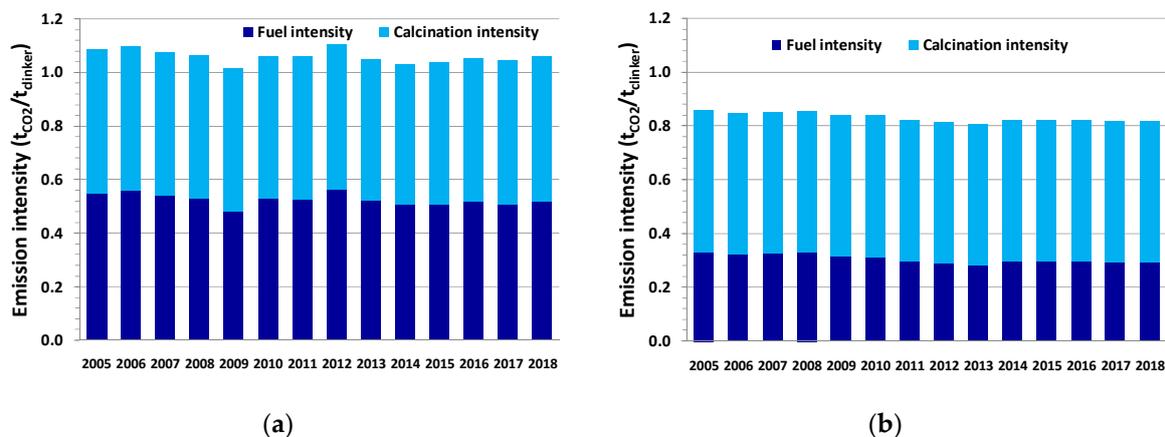


Figure 1. Carbon dioxide emission intensity in the Spanish cement industry from 2005 to 2018: (a) for white clinker; (b) for gray clinker.

The total emission intensity of the Spanish cement industry decreased from 0.86 to 0.83 t CO₂/t clinker for a period of time from 2005 to 2018 (Figure 2), similarly to the EU28 cement industry (from 0.87 to 0.81 t CO₂/t clinker in the same period) as shown in Figure 3. However, recent reports have suggested that lower values for the total carbon dioxide emission intensity should be achieved in the next few years [8,27,28].

The Taxonomy Technical Report, developed by the Technical Expert Group on Sustainable Finance, a European Union classification system for environmentally sustainable economic activities (Taxonomy), was presented in 2019 [27]. The Taxonomy sets a performance threshold of 0.766 t CO_{2e}/t clinker applicable to cement clinker factories, whereas a performance threshold of 0.498 t CO_{2e}/t cement is the upper limit for cement factories. The first upper limit is based on the EU ETS benchmark for gray cement clinker because it corresponds to the performance level reached by the 10% most efficient cement plants in the European Union (EU) [28] and the second one is estimated considering a threshold for the clinker factor (clinker/cement ratio) of 0.65 (0.766 × 0.65 = 0.498 t CO_{2e}/t cement).

Furthermore, the EU’s top 10% of most efficient Portland cement installations could reduce the total emission intensity up to 720 kg CO₂/t clinker for gray cement clinker and up to 965 kg CO₂/t clinker for white cement clinker by the end of the second decade of the twenty-first century. Therefore, in order to achieve total carbon dioxide emission intensity values lower than 0.766 t CO₂/t clinker, which entail a reduction of the current value by 7.7%, the Spanish cement industry will implement measures in the following areas: alternative raw materials, energy and alternative fuels (biomass and others), low-carbon clinkers, and carbon dioxide capture.

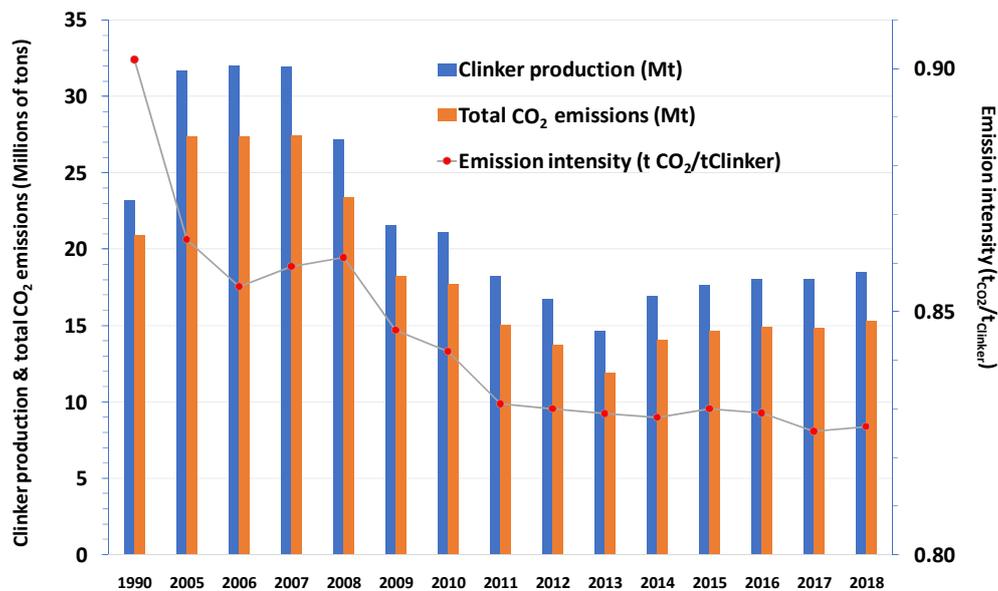


Figure 2. Comparison between the total emission intensity in the Spanish cement industry from 2005 to 2018 plus 1990 and the clinker production and total CO₂ emissions.

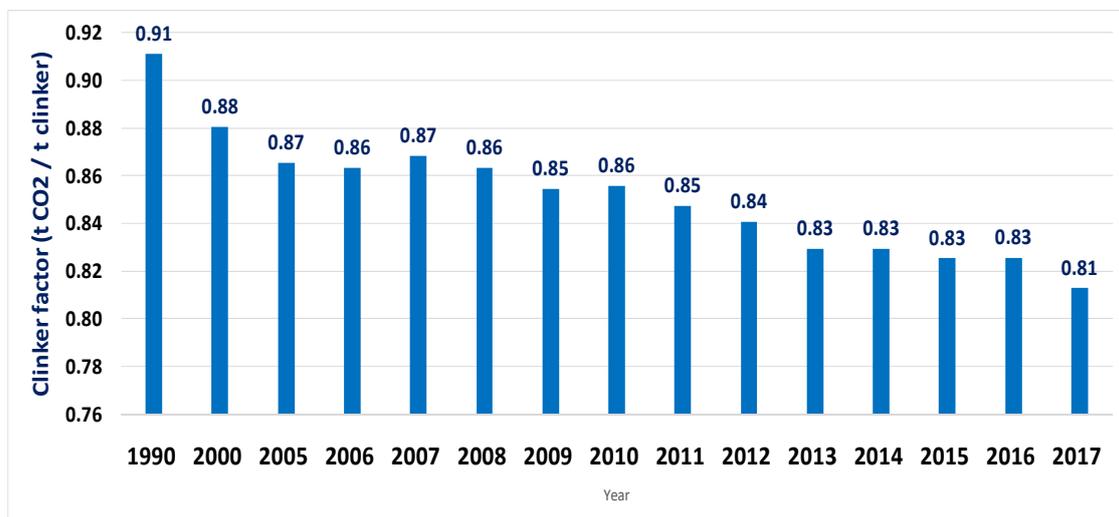


Figure 3. Clinker factor in the group of 28 countries of the European Union (EU) known as EU28 cement industry from 2005 to 2017 plus 1990 and 2000 [29].

3.1.1. Alternative Raw Materials

The alternative raw materials use in Spain is growing slowly but steadily (Figure 4). The alternative raw materials rate in 2018 was 3.1%, but it is expected to be about 5% in 2030 and 8% (−25 kg CO₂/t cement) in 2050. For instance, to achieve these targets, the Spanish cement industry assumes to increase the recycled concrete up to 10% in 2030 and up to 20% in 2050.

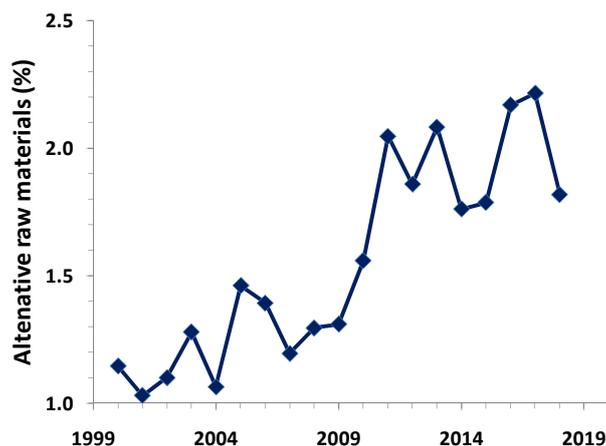


Figure 4. Alternative raw materials consumption by the Spanish cement industry from 2000 to 2018.

3.1.2. Energy and Alternative Fuels

European energy policies for carbon dioxide emission reduction stimulate to achieve certain goals by reducing the share of fossil-fuels consumption, the demand for energy and the thermal and electric energy intensity.

Improved energy efficiency and fuel-switching for industrial use, for example from fossil fuels to biomass and waste materials, represents an opportunity for the cement industry to achieve net-zero carbon emissions by 2050. In order to improve the energy efficiency, it will be necessary to reduce the thermal energy intensity of clinker production and the electric intensity of cement grinding by retrofitting existing kilns and mills.

It is well-known that clinkerization process has been used since many decades to burn a wide range of waste materials. This burning process provides good conditions, i.e., high temperature and firing under oxidizing atmospheres, for using a wide range of wastes. In particular, the use of biomass as fuel in cement clinker kilns can reduce significantly carbon intensity of cement clinker.

Alternative fuel rate evolution in Spain from 2000 to 2018 is shown in Figure 5. Rate evolution was less impressive for the first decade of the 21st century, from 0.9% to 15.8%, than for the second one, where 26.5% was achieved in 2018. Figure 6 shows the avoided CO₂ emission by biomass in the Spanish cement industry from 2005 to 2018. On the other hand, with the continued maturing of the renewable energy industry in Europe, it is expected to reach net-zero carbon emissions in the electrical sector in a short period of time.

In the European cement industry, it is expected to meet the thermal energy intensity range of 2.9–3.4 GJ/t clinker according to the Taxonomy document mentioned above [27]. The European Cement Association expects to achieve a 16% efficiency improvement in thermal energy from 2018 to 2050, i.e., fuel efficiency from 3.6 to 3.0 GJ/t clinker.

With regard to the electrical energy consumption in cement factories, mills, and exhaust fans consume 80% of the total amount of energy (90–150 kWh/t cement [30,31]). Then, a significant part of the decarbonization of the cement industry will go in parallel with the energy sector decarbonization (electricity produced from renewable sources). In addition, new technologies will allow achieving an average electricity consumption of 85 kWh/t cement. Included among these are new vertical roller mill designs for raw, fuel, and cement grinding with integral high-efficiency separators. In addition, the production and use of new blended Portland cements (ternary and quaternary) with lower clinker factors is an effective route of reducing energy consumption.

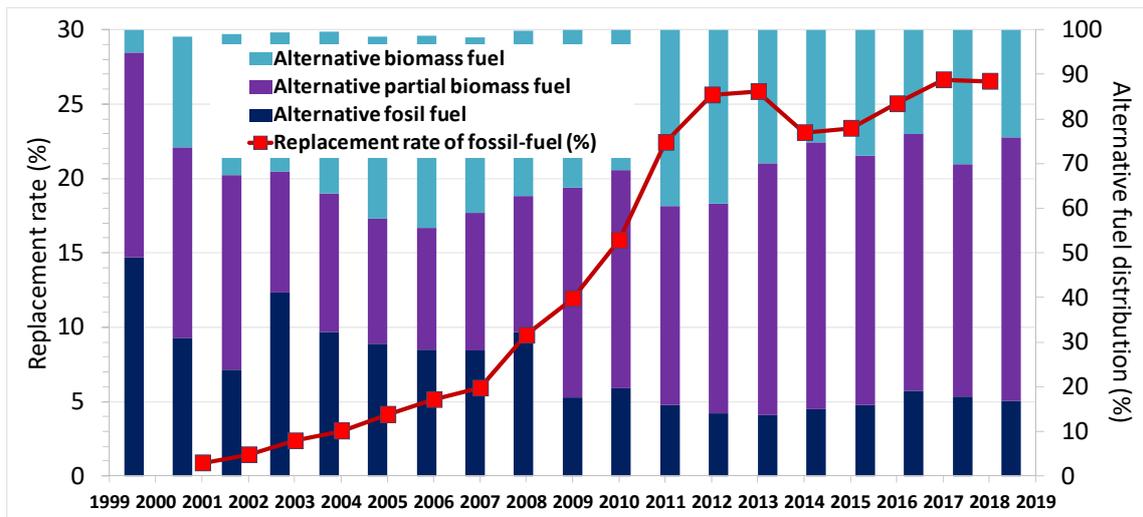


Figure 5. Evolution of the replacement rate of old fossil-fuels and alternative fuel distribution by the Spanish cement industry.

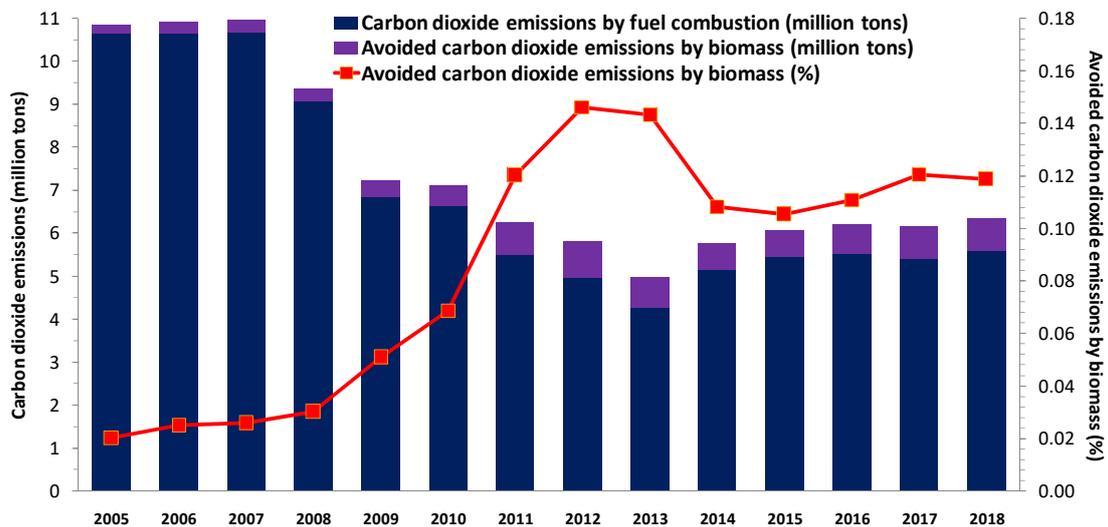


Figure 6. Carbon dioxide combustion emissions and avoided CO₂ emission by biomass in the Spanish cement industry from 2005 to 2018 (millions of tons).

Summing up, one adequate alternative measure, with regard to the energy consumption in Portland cement clinker factories, is the fossil fuel switching. In addition, improving the energy efficiency and the potential electrification of the clinker kiln should be investigated.

The Spanish cement industry urgently need to reduce the burning of fossil fuels and increase in thermal efficiency. It is expected a drawdown of carbon dioxide emissions due to the use of 20% biomass fuels in 2030, followed by a 40% (−79 kg CO₂/t cement) in 2050. These figures have been calculated after deducting the improvement in thermal efficiency which is predicted to be 5.5% in 2030 and 16% (−39 kg CO₂/t cement) in 2050 over the year 2018 (clinkerization process and waste heat recovery). Given that, the thermal energy intensity will decrease from 3.603 GJ/t clinker (2018) to 3.4 GJ/t clinker in 2030 and to 3.0 GJ/t clinker in 2050. Consequently, it can be assumed a rate of decline about 0.2 GJ/t clinker every ten years. An additional 10% of carbon dioxide emission reduction from fossil-fuels combustion can be achieved in 2050 by using new sources of energy, i.e., hydrogen and the potential electrification of the clinkerization process (−20 kg CO₂/t cement).

3.1.3. Alternative Clinkers

Belite and sulfoaluminat clinkers [32,33], among others, need less calcium carbonate in the raw materials; therefore, they emit less carbon dioxide compared to the current Portland cement clinker. Their availability is limited in Spain. So far, the Spanish Code on Structural Concrete (EHE-08) [34] does not include cements made with such clinkers. Further studies are required to assess the durable performance of the potential alternative clinkers. Later on, it would be necessary to develop and implement technical standards based on the consensus of the different stakeholders.

By providing an additional source of low-carbon clinkers and a route into standardization and market acceptance, such binders are particularly relevant. Thus, the Spanish cement industry assumes a 2% of reduction of the process emissions in 2030 and 5% (−18 kg CO₂/t cement) in 2050, after deducting the carbon dioxide taken off by the recycled concrete re-use, by considering these low-carbon clinkers.

3.1.4. Carbon Capture, Utilization, and Storage (CCUS)

Carbon dioxide capture, utilization, and storage (CCUS) technologies will provide the direct mitigation of process and fossil emissions in the cement sector. Recently, the Technical Committee ISO/TC 265/WG 1 “Carbon dioxide capture, transportation, and geological storage” belonging to the International Organization for Standardization (ISO) has developed a study on carbon dioxide capture technologies in the cement industry [35]. Such report updates former studies on carbon dioxide capture and utilization technologies [36–38]. Some technologies for carbon dioxide capture from cement factories are under development. For instance, a new post-combustion capture technology is based on amine absorption, where the carbon dioxide from the exhaust gas is, firstly, absorbed in a circulating aqueous amine solution, and after, regenerated with steam treatment [38].

These breakthrough technologies in carbon capture and utilization (CCU) and carbon dioxide carbon capture and sequestration (CCS) that are expected to remove this gas that contributes to global warming will be the last step used by the cement industry to guarantee the net zero carbon footprint in 2050. According to the calculations presented in this paper, the Spanish cement would need to capture 272 kg CO₂/t cement by 2050. Therefore, CCS and CCU are two key technologies for the decarbonization of Europe. In short, it can be said that all the clinker measures together, without CCS, will offer a cut-down on the carbon dioxide emissions of 79 kg CO₂/t cement (1%) in 2030 and 181 kg CO₂/t cement in 2050. In addition, carbon capture and storage (CCS) technologies should provide a reduction of 272 kg CO₂/t cement (50%) by 2050.

3.1.5. Mortar and Concrete Carbonation

Finally, the carbon dioxide released during the calcination process can be reabsorbed by cement-based materials in different amounts as shown in Tables 1 and 2. Recently, a range between 15–20% of the carbon dioxide released during the calcination process has been suggested [13].

(Re-)carbonation is assumed to be 20% IPCC reported calcination emissions in 2030 (−73 kg CO₂/t cement) and 23% in 2050 (−72 kg CO₂/t cement). This additional 3% is due to the expected improvement in the carbonation techniques applied to the recycled concrete [14].

This aspect has been addressed in its fullest extent in the fourth part of this paper.

3.2. Cement Measures

3.2.1. Clinker Factor

Clinker is the main Portland cement constituent. When finely ground and mixed with gypsum, it reacts in contact with water and forms a fluid paste which sets and hardens. In addition, some mineral cement constituents such as siliceous fly ash, natural pozzolan, and ground granulated blast-furnace slag, among others, have pozzolanic characteristics. These can be utilized to partially replace clinker in Portland cement, thereby reducing the amount of clinker per ton of cement and the fossil-fuel and carbon dioxide emissions associated with Portland cement clinker production.

Reducing the clinker to cement ratio has been a significant way for the decarbonization in Spain along the time. The clinker factor decreased from 0.812 in 2000 up to 0.772 in 2007. In that period of time, the cement consumption was growing constantly in Spain, while the clinker factor was declining. Then, it increased continuously from 2008 (0.773) to 2018 (0.828) by almost 7 percentage points (Figure 7). This is a negative development and hopefully this trend will change in the next years.

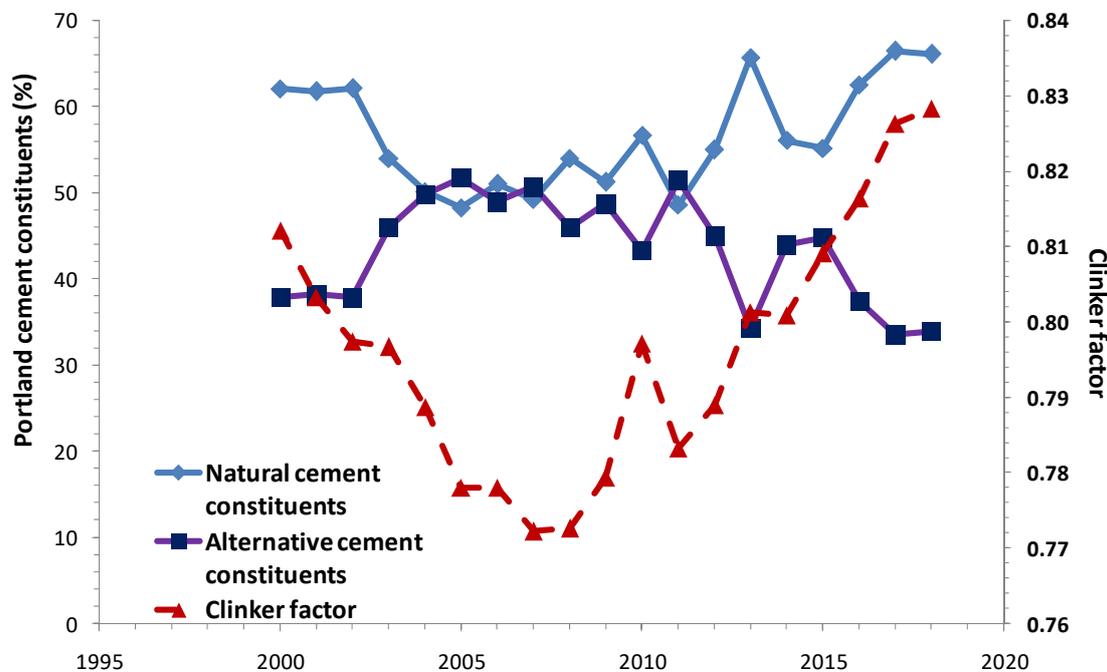


Figure 7. Clinker factor and alternative cement constituents in Spain from 2000 to 2018.

The target value for the Spanish cement industry is to decrease the clinker content from 83% in 2018 till 75% by 2030 (Drop-off in carbon dioxide emissions: 70 kg CO₂/t cement) and 70% in 2050 (drop-off in carbon dioxide emissions: 114 kg CO₂/t cement); whereas for the European Cement Association, CEMBUREAU, the target is to decrease the clinker content from 77% to 74% in 2030 and 65% in 2050 [39]. In both cases, these goals will entail a reduction of the current value by 16% within the next thirty years. This lever should be promoted by providing an adequate route into standardization of ternary and quaternary Portland cements among other types of blended cements. Furthermore, the market acceptance of the mentioned cements and standards is undisputed.

In addition, the share of blended cement production in Spain was 67% in 1995, which increased to 72% in 2000 and 78% in 2005 [40,41], then, it remained almost same in 2009. Nevertheless, this value has fallen from 75% (2010) to 63% (2016) and the carbon dioxide emission intensity for Spanish cements increased from 711 kg CO₂/t cement to 757 kg CO₂/t cement over the same period of time [41]. It is less than Spanish cement industry would have wished and it should be less than The Ministry for Ecological Transition and Demographic Challenge (MITECO) would have expected [42]. Consequently, the deployment of blended Portland cements amongst Portland cement users should be stimulated.

Figure 8 is presented for comparison purposes. There was a 5% decrease of the direct carbon dioxide emission intensity from 2010 to 2017 amongst Cement Sustainability Initiative (CSI) and non-CSI members [29]. By contrast, the direct CO₂ emissions intensity with regard to the cement production (kg CO₂/t cement) increased in Spain from 2010 to 2015. The percentage of blended cement is decreasing in Spain due to the quite low differences in cost of some blended cements and CEM I without additions, i.e., currently, the €/MPa is lower in CEM I than in blended cements. This trend must be reversed in the next years.

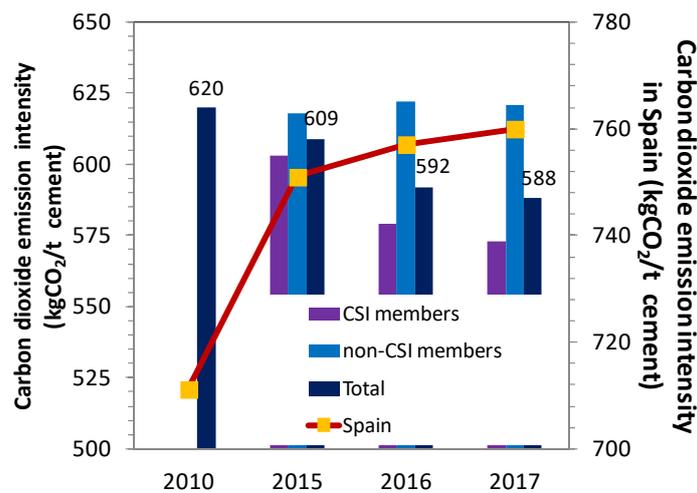


Figure 8. Direct carbon dioxide emission intensity, i.e., emissions from sources controlled by the reporting cement factory, such as emissions from kiln fuels related to clinker production, calcination of carbonates, and combustion of organic carbon from raw materials, for the Cement Sustainability Initiative (CSI) and non-CSI members (kg CO₂/t cement).

3.2.2. Cement Grinding

Regarding the carbon-neutral renewable energy use in cement plants, it is assumed that at least 50% of renewable electricity will be used by 2030. Furthermore, it is expected that 100% of the electricity will be renewable by 2050. Renewable energy technologies, which range from wind, solar photovoltaic, solar thermal, hydroelectric, tidal, geothermal to biomass, do not use fossil fuels. They have zero greenhouse gas emissions. If the Spain's Integrated Energy and Climate Program (2021–2030) is met [42], around 50% of the Spain's electricity will be provided from renewable energy sources by 2030 and 100% by 2050, compared to today (37.5%). The carbon dioxide reduction due to this lever will be of about 114 kg CO₂/t cement in 2050.

On the other hand, the energy will be 100% electricity generated from renewable sources and hydrogen in 2050 (Drop-off in carbon dioxide emissions: 36 kg CO₂/t cement) and is expected to be 50% in 2030 (Drop-off in carbon dioxide emissions: 18 kg CO₂/t cement). Furthermore, the use of local raw materials minimizes energy consumed by transport. Besides, carbon-neutral road transport is possible by 2050 (hydrogen and electric vehicles, EV) with a drop-off in carbon dioxide emissions: 9 kg CO₂/t cement.

Summing up, considering all the cement measures together, the carbon dioxide emissions will be cut-down by 88 kg CO₂/t cement in 2030 and about 159 kg CO₂/t cement in 2050.

3.3. Concrete Measures

Only a few scenarios of improvement or potential levers have been considered. We have done a great deal of work to put a good proposal on the table, which can be summarized as follows:

- Improving the concrete mix design to enhance the quality of the final material
- Use of high-performance concrete
- Increasing the low-carbon cement content (considering circularity in the cement production) for the same durability and strength by an adequate selection of all the constituents, particularly the adequate use of additives
- Development of new mix designs at the concrete scale including recycling strategies
- Increasing the use of pre-cast elements
- Reducing waste in construction
- Post-tensioning for increased strength

- Optimizing the structural design to limit over-specification
- Implementing breakthrough construction techniques including 3D-printing

Together these actions represent a significant move to address the carbon dioxide reduction in 2030 (5%) and 2050 (10%). The carbon dioxide emissions will be cut-down by 28 kg CO₂/t cement in 2030 and about 40 kg CO₂/t cement in 2050. Furthermore, carbon neutral transport will provide additional 5 kg CO₂/t cement by 2050.

4. Carbonation of the Spanish Cement-Based Materials

A preliminary study performed by the IETcc commissioned by the Spanish cement industry showed that the cement-based materials are able to uptake at least 3% of the carbon dioxide emitted for the calcination process, when a low degree of carbonation is considered (DoC = 15–20%) [15]. Later on, it was realized that the actual degree of carbonation was much higher (DoC = 62.5%) and, therefore, the carbon dioxide uptake was updated (11%) to take into account the surface area to volume ratio (S/V) of three [12]. However, some precast elements have higher ratios (S/V = 8 for 0.5 × 0.5 m concrete pillars; S/V = 20 for façade panels). By contrary, some building structures and civil works present ratios as low as the unity. In addition, carbonation of concrete foundations can be considered to be negligible in many cases.

In order to quantify the carbon dioxide uptake (ACDU) by mortars and concretes, a simplified method or Tier 1 can be used [13,14]. This methodology shall consist of a system for measuring the carbon dioxide uptake during the service-life and during the end-of-life and secondary use according to Equations (1) and (2), respectively. This method has been pursued for the cements produced in Spain from 1898 to 2018. The alpha and beta parameters were 0.20 and 0.03 for the present study [13]. It should be mentioned that this Tier 1 does not consider the final use of the Portland cements.

Cement production grew in Spain continuously from 1898 to 1978 (except 1936–1939). In 1979, it followed a decreasing trend up to 1986 and increased slightly during the next years. From 1990 to 1993, it decreased again, and then, it increased sharply from 1993 (23.9 million tons) to 2006 (47.5 million tons). Then, it falls down until 2016 (13.7 million tons). This trend was reversed from 2017 (Figure 9). As expected, Portland cement production is linked not only to the carbon dioxide emissions, but also to the carbon dioxide uptake. Therefore, similar trends are observed.

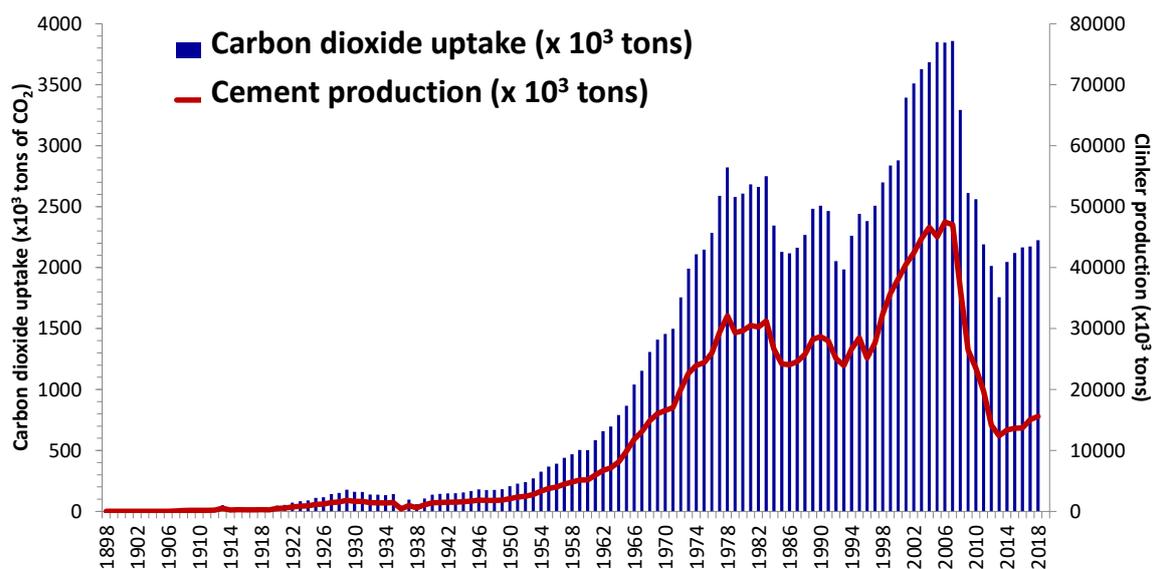


Figure 9. Portland cement production and estimated carbon dioxide uptake by using the Tier 1 approach [13] in Spain from 1898 to 2018 (120 years).

Carbon dioxide uptake due to the carbonation of mortars and concretes made with Portland cements manufactured in Spain has been estimated from 1898 to 2018 (Figure 9). The assumptions at the basis of this study are as follows:

- Carbon dioxide emission data, from 2005 to 2018, are those reported in yearly greenhouse gas inventories from Parties to the United Nations under the authority of the United Nations Framework Convention on Climate Change (UNFCCC). From 1992 to 2015, they were estimated from actual clinker production data collected by Oficemen [43]. Finally, carbon dioxide emission data in the period of time between 1898 and 1991 were estimated from Portland cement production data collected from different sources [44,45] in Spain
- As a preliminary hypothesis it was postulated that the only type of cement from 1898 to 1964 was CEM I (cement made by grinding clinker and 6% of gypsum). Thereafter, the clinker factor was used to calculate the clinker production from the cement production (1965–1991)
- The calcination carbon dioxide emission intensity value was assumed to be 0.52 in the period of time between 1898 and 2004
- Carbonation uptake was estimated from 1898 to 2000 taken into account only the service-life according to Equation (1). In addition, the end-of-life and secondary use were also considered thereafter following Equation (2)

As result of the study carried out in this study, the total carbon dioxide uptake can be assumed to be 0.14 Gt from 1898 to 2018 (120 years) by using the simplified method or Tier 1 suggested elsewhere [13]. Only 10.8% of the total carbon dioxide absorption was produced during the first seventy years, i.e., from 1898 to 1968. By contrast, 77% of the carbon dioxide uptake was attributed to the clinker manufactured over the last forty years, i.e., from 1978 to 2018, due to the greater Portland cement demand in this period of time.

Xi et al. [21] performed a global study on CO₂ uptake by carbonation of cement-based materials between 1930 and 2013. They found that a cumulative amount of 4.5 Gt C (16.5 Gt CO₂) was uptake by carbonation of cement-based materials, i.e., 43% of the calcination carbon dioxide emissions (38.2 Gt CO₂ released by calcination from 1930 to 2013).

In this study, a cumulative amount of 0.13 Gt CO₂ is estimated to be sequestered by carbonation of mortars and concretes made with Spanish cements between 1930 and 2013. Accordingly, the Spanish contribution to the carbon dioxide offsetting in that period is only 0.78% with regard to the global CO₂ uptake.

In short, cement-based materials are carbon dioxide sinks, which are not currently considered in the GHGs emissions inventories, but that is what they are.

5. Summary of the Technical Pathways for the Spanish Cement Sector

The Spanish cement industry is in the race to become climate-neutral in 2050, and the contribution of different levers to emission intensity reduction was presented in previous parts of this paper. All of them have now been compiled to show the path to achieve the net zero greenhouse gas emissions [46]. Figure 10 shows a tentative waterfall chart summarizing a proposal of technical pathways for the Spanish cement sector of reducing its carbon footprint to net zero. It should be mentioned that transportation emissions are not considered as they represent only a low percentage of the total carbon dioxide emissions of Portland cement production. It has not been considered, either, to what extent concrete efficiency in use might result in a decrease in carbon dioxide emissions.

As part of this research, the changes in carbon dioxide emission intensity and the contributions from different levers have been analyzed. The direct carbon dioxide emissions intensity has gone down by 86 kg CO₂/t cement to 729 kg CO₂/t cement in 2018 as compared to the baseline year (1990). Waterfall chart shown in Figure 10 collects the carbon dioxide reductions proposed by the Spanish cement industry for a technical transition to achieve the carbon neutrality by 2050. All the levers or

scenarios of improvement in the reduction of CO₂ emissions are grouped into five categories, and each one is named by a letter “C” (5Cs): clinker, cement, concrete, construction, carbonation [39].

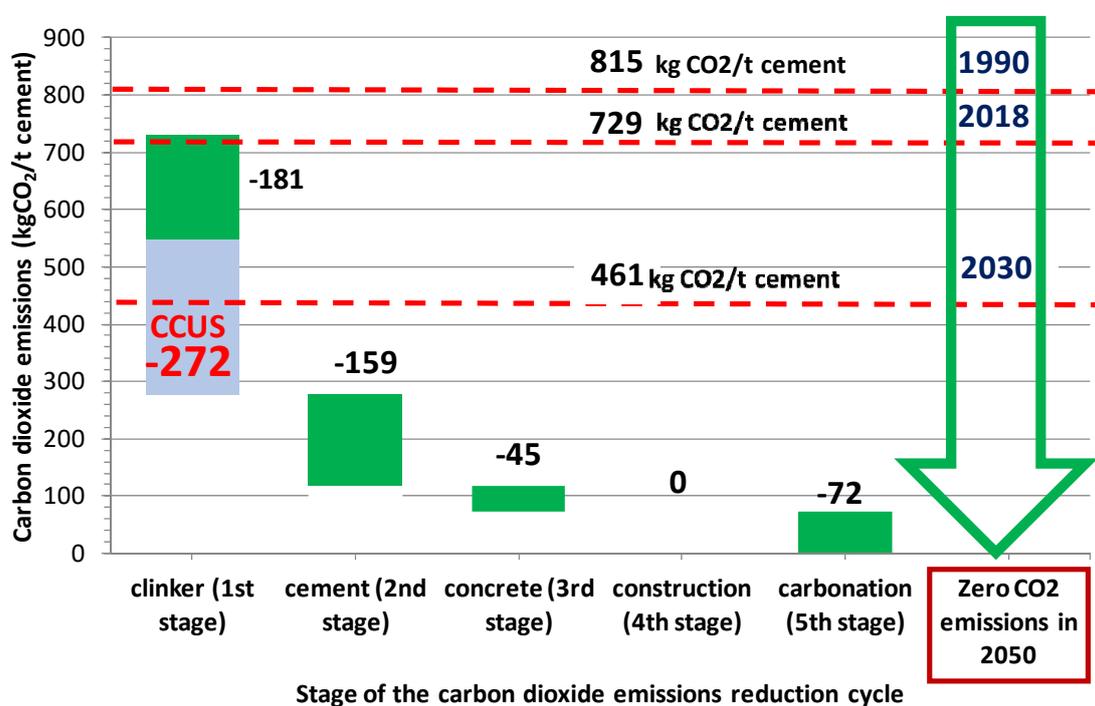


Figure 10. Waterfall chart summarizing the transition from 2018 to 2050 to achieve the carbon neutrality for the Spanish cement industry. These values represent the drop-off in carbon dioxide emissions for each stage considered: clinker, cement, concrete, construction, and carbonation (5Cs).

The starting point in Figure 10 are the direct and indirect emissions released in 2018 (729 kg CO₂/t cement). Then, the carbon dioxide drop off expected for each stage in 2050 is shown: clinker (−181 kg CO₂/t cement), cement (−159 kg CO₂/t cement), concrete (−45 kg CO₂/t cement), construction (−0 kg CO₂/t cement), carbonation (−72 kg CO₂/t cement). In summary, the existing mitigation technologies related to the clinker, cement and concrete production and use are insufficient to reach the expected carbon neutrality, i.e., the net zero carbon footprint. Accordingly, nascent and breakthrough technologies such as novel advanced carbon dioxide capture, utilization and storage (CCUS) technologies should be implemented by the Spanish cement sector to ensure the climate neutrality (−272 kg CO₂/t cement).

Zero value for construction means that Spanish cement industry does not take into account carbon dioxide drop off due to this stage. Nevertheless, The European Cement Association (CEMBUREAU) considers carbon dioxide drop off due to concrete in use of −54 kg CO₂/t cement in 2030 and −89 kg CO₂/t cement in 2050 [39].

This approach considers that 43% of the carbon dioxide released in 1990 (815 kg CO₂/t cement) will be reduced in 2030 (461 kg CO₂/t cement). However, The European Union (EU) has proposed a new target with regard to the greenhouse gas (GHG) emissions reduction of at least 50%–55% for 2030 rather than 40%, from 1990 levels [47], but carbon neutrality will be reached by 2050 anyhow. The commitment of the Spanish cement industry to achieve a neutral balance between carbon dioxide emissions and carbon dioxide sequestration will be arranged between now and 2050 combining different lever’s contribution.

6. Conclusions

This paper outlines different pathways towards net-zero carbon dioxide emissions to achieve the European Green Deal aim. Particularly, this target will be reached by balancing carbon dioxide

emissions with carbon dioxide removal through mortar and concrete carbonation. In addition to this carbon dioxide sink, several potential levers or scenarios of improvement in the reduction of carbon dioxide emissions for the Spanish cement industry have been presented in this paper. They have been defined in every stage of clinker, cement and concrete production and use.

In conclusion, the net carbon dioxide emissions of the Spanish cement industry (carbon dioxide emissions produced during the calcination process less the carbon dioxide uptake by mortars and concretes) will be implemented in the Roadmap 2050 of the Spanish cement industry, though this carbon sink is not currently considered in emissions inventories. Hence, carbon dioxide uptake by cement-based materials should be included in the climatic models of the future IPCC's Assessment Reports to minimize the gaps found in such models. Furthermore, the National Climate Change Offices should implement simplified and advanced methodologies to calculate the net carbon dioxide emissions in the National Greenhouse Gas Inventories.

A global estimation has been performed to compute the carbon dioxide uptake by the Portland cements produced in Spain from 1898 to 2018. Accordingly, the simplified methodology or Tier 1 considers an average carbon dioxide uptake of 20% of the value of the emitted calcination carbon dioxide during the service life and 3% for the end-of-life and secondary use. The whole carbon dioxide uptake by carbonation of cement-based materials was deemed to be 0.14 Gt (120 years). Most of this amount (77%) is allocated to the cements manufactured during the last forty years (1978 to 2018).

Finally, carbon dioxide capture, utilization and storage (CCUS) technologies should be implemented by the Spanish cement industry to achieve zero carbon dioxide emissions by 2050. The commitment of the Spanish cement industry to achieve a neutral balance between carbon dioxide emissions and carbon dioxide sequestration will be arranged between now and 2050.

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