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Natural and enhanced carbonation of lime in its different applications: a review

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ABSTRACT

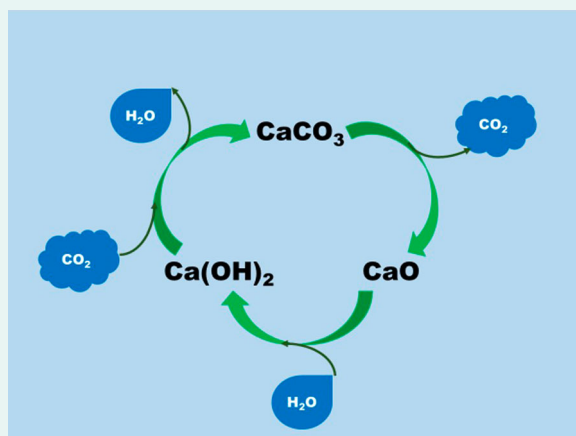
Lime is a product derived from the thermal decomposition of limestone (mainly calcium carbonate, CaCO_3) into quicklime (CaO) and carbon dioxide (CO_2), also called calcination. Controlled reaction with water is used to manufacture hydrated lime (Ca(OH)_2) products. Lime is used in a wide variety of applications: metals industry, construction materials sector, civil engineering, environmental protection, agriculture, and chemical industry. Lime production is one of the sources of anthropogenic CO_2 emissions resulting in global warming and ocean acidification. However, a proportion of the CO_2 emitted during the calcination is reabsorbed by the lime during the product life within its different applications. This process called carbonation is thermodynamically favoured because it is exothermic. It allows permanent CO_2 storage in a stable product since the lime combines with gaseous CO_2 reforming CaCO_3 . This paper reports a comprehensive literature review on the carbonation potential of lime in different applications. The total carbonation potential is assessed as carbonation rate, i.e. the ratio between the CO_2 reabsorbed through carbonation during the operational life of lime and the CO_2 emitted during limestone calcination. This study provided preliminary evidence that, based on the current EU market, on average 23–33% of lime process emissions are carbonated during the use phase. Carbonation over time is also analysed for the lime applications where information is available. For three applications, namely water, flue gas cleaning and pulp and paper, the carbonation reaction is instantaneous. Up to 22% of the calcination emissions are absorbed within five years based on the current EU market.

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Quicklime; hydrated lime; carbonation; carbon dioxide; carbon sink




1. Introduction

The process emissions of lime production, i.e. the CO_2 released during limestone calcination excluding that released from the combustion of fossil fuels, are estimated to account for about 1% of the global anthropogenic CO_2 emissions excluding those associated to

land use change [1]. Anthropogenic CO_2 emissions are the accepted cause of global warming and ocean acidification [2]. Limestone calcination is defined as the thermal decomposition of limestone, i.e. the transformation of calcium carbonate (CaCO_3) into quicklime (calcium oxide, CaO) and carbon

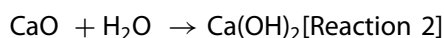
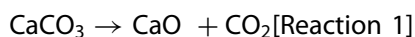
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dioxide (CO₂) as shown in Reaction 1. Quicklime can then be mixed with water to produce hydrated lime Ca(OH)₂, according to Reaction 2, which is beside quicklime a widely used product. Hydrated lime is applied in a wide range of sectors, from the metals industry to the construction materials sector, in civil engineering, environmental protection, agriculture, and in the chemical industry. During the operational life of products containing lime, the hydrated lime will react with atmospheric CO₂ according to Reaction 3, generating calcium carbonate again, thus removing CO₂ from the atmosphere and storing it permanently [3, 4].



The carbonation reaction is exothermal and therefore thermodynamically favourable. Lime carbonation can occur in natural conditions (i.e. under ambient carbon dioxide concentrations) and in enhanced conditions (i.e. under enhanced carbon dioxide

concentrations and by optimising other parameters such as the temperature and the relative humidity). Enhanced carbonation conditions help to accelerate the process. The routes for accelerating carbonation can be direct or indirect. In the direct process, carbonation occurs in a single step, while in the indirect one the lime is initially extracted from the mineral matrix and subsequently carbonated [5].

The scope of the present work is to assess the carbonation potential of lime in its different applications by reviewing the available literature on the topic, as well as the evolution of carbonation over time for the lime applications where information is available.

2. Materials and methods

The literature assessment about the carbonation potential of lime during its use phase has covered almost all its market sectors, in particular the ferrous (iron and steel) industry, the construction materials (sand lime bricks (SLB), light-weight lime concrete: autoclaved aerated concrete (AAC), mortars, hemp-lime construction materials), civil engineering (soil stabilisation and asphalt pavements), environmental protection (flue gas purification, treatment of drinking water, wastewater, biosolids and dredging sediments), agriculture, the chemical industry, and pulp and paper applications and other industries (typically in the non-ferrous metals sector). The analysed market sectors and applications with their respective market shares for the European Union including the United Kingdom in 2018 are reported in Table 1 according to data provided by the European Lime Association (EuLA).

For each application, a review of the potential carbonation rate of lime was performed, in terms of natural as well as enhanced carbonation (the latter only for the applications where it is actually undertaken). The assessment was mainly based on peer-reviewed papers, on technical reports from research or institutional bodies (such as the Joint Research Centre or Environmental Protection Agencies), on the Best Available Techniques Reference (BREFs) Documents, on patent documents, on Master and Doctoral theses, and on industrial company websites.

The applications are grouped based on the robustness of the information about carbonation found in the assessed literature in the following clusters:

Table 1. Lime market sectors and applications analysed in this study with their respective market shares, according to data provided by EuLA for the European Union including United Kingdom in 2018.

Lime market sector	Lime application	Market share
Iron & Steel industry	Iron & Steel industry	41%
Construction	Sand Lime Brick (SLB)	2.9%
Materials	Light-weight lime concrete:	4.4%
	Autoclaved Aerated Concrete (AAC)	
	Pure air-lime mortars	0.4%
	Mixed air-lime mortars	2.9%
	Hemp-lime	0.1%
	Other (paints, natural fibres ...)	1.6%
Civil Engineering	Soil stabilisation	6.4%
	Asphalt pavements	0.1%
Environmental Protection	Drinking water	2.1%
	Wastewater/ Biosolids treatment	4.3%
	Sludge/Dredging sediments treatment	2.9%
	Flue gas purification systems	7.8%
	Other (Acid Mine Drainage – AMD, lake liming ...)	0.6%
Agriculture	Fertiliser	1.6%
	Other (sanitation, aquafarming, ...)	1.4%
Chemical Industry	Calcium carbide	0.3%
	Soda	0.1%
	Petrochemical	1.6%
	Other (salts, leather tanning, ...)	5.0%
Other Industrial Consumers	Sugar	0.2%
	Glass	0.8%
	Non-ferrous metal industry without aluminium	1.5%
	Aluminium	1.4%
Pulp & Paper	Other	3.0%
	Pulp and Paper	5.9%

- Applications with conclusive scientific data, were defined those where the amount of literature published on carbonation allows for statistical treatment or the results presented are consistent throughout these publications/reports to establish the carbonation rate being natural or enhanced.
- Applications with less conclusive scientific data, where the amount of literature published on carbonation is very insufficient to allow data comparability, thus the outcome is less conclusive to establish any carbonation rates (natural or enhanced).
- Applications with no information available, where due to the low volume of lime in the application there are few studies on carbonation, and these limited studies provide no estimate the carbonation rate.
- Not assessed applications, where due to the low quantities of lime in the application, there are no dedicated studies on carbonation. Thus, they are not covered in the scope of this literature review.

The carbonation potential of each application was assessed as the carbonation rate, i.e. the ratio between the amount of CO₂ absorbed during carbonation and the amount emitted during calcination. In the applications where lime is fully carbonated, the carbonation rate is 100% and the lime application can be considered carbon neutral since the CO₂ emitted during calcination, i.e. excluding the emission from fuel combustion, is completely reabsorbed. The process CO₂ emissions coming entirely from lime calcination was assumed equal to 786 kg CO₂ per tonne of lime, according to the Best Available Techniques Reference (BREF) Document for Cement, Lime and Magnesium Oxide Manufacturing Industries published by the European Commission pursuant to Emissions Directive 2010/75/EU (Integrated Pollution Prevention and Control) [6].

3. Results

The assessment was based on a total of 557 documents listed in Supplementary Materials 1 and 2, of which 224 provide quantitative and qualitative information useful for the evaluation of the carbonation rate in the specific lime applications listed in Supplementary Material 1. Table 2 shows the main characteristics of the relevant documents, most of them are peer-reviewed papers and the Best Available

Techniques Reference documents (BREFs), which account for around 80% of the assessed literature. Most studies especially the ones dealing with enhanced carbonation, addressed carbonation at the laboratory or at the pilot scale. Generally, European literature sources are available for most applications, except for aluminium production, titanium dioxide production and Autoclaved aerated concrete (AAC), for which mainly non-EU data sources were identified.

Table 3 reports the main literature findings about the carbonation rates in the different applications where lime is used. Each carbonation rate is expressed as the percentage of the CO₂ emitted during the production of lime, excluding the fuel combustion.

3.1. Lime applications with conclusive scientific data on carbonation

About 61% market share for lime (according to Table 1) have relevant and consistent information in the literature allowing conclusive assessment of natural and enhanced carbonation rates for these applications, as summarised in Table 3.

Based on the factors which affect enhanced carbonation reported the assessed literature, the conditions for effective enhanced carbonation in conclusive applications is reported in Table 4.

Steel slags is the major application of lime, as shown in Table 1. The presence of Ca-compounds in the steel slag is the consequence of using quicklime (CaO) or limestone (CaCO₃) during the iron and steel making processes. Quicklime is used in the hot metal desulphurisation processes as well as in the Basic Oxygen Furnace (BOF) and Electric Arc Furnace (EAF) processes, as a fluxing agent to create a basic slag that can neutralise acid-forming elements, remove sulphur, phosphorous, silica and alumina inclusions and protect the refractories [7]. Lime is also used in a broad variety of secondary metallurgical processes for the removal of additional impurities and to prevent the reabsorption of impurities from the slag. In addition, quicklime can be used together with other materials, such as fluorspar, to form a synthetic slag which is used as a flux to remove additional sulphur and phosphorus after the initial steel refining process. In the pig iron production process, quicklime is used far less, where it is mixed with limestone in a proportion of 1:6, mainly in the sintering process [7].

Table 2. Main information about the literature assessment: number of papers consulted per each application and relative characteristics.

Lime market sector	Lime application	N° total sources	Type of source	Type of test (field vs. lab test)	Test duration	Geographical context
<i>Iron and steel industry</i>	Iron slags	NC: 2	Peer-reviewed papers	Field tests	6 months - 100 years	1 paper: United Kingdom; 1 paper: U.S.A.
		EC:12	9 peer-reviewed papers + 1 BREF document + 2 websites (statistics about European steel and slags production)	Lab-tests	Generally 1 h	4 papers: Europe (United Kingdom, Belgium, Poland, and Finland) 5 papers: Korea, Taiwan, China
	Steel slags	NC: 8	5 peer-reviewed papers + 1 BREF document + 2 websites	3 papers: field tests 1 paper: lab tests 1 paper: both cases	Lab-scale: 1–12 weeks Field cases: 4–18 months	3 papers: Europe (Italy, Germany, and Spain) 2 papers: U.S.A.
		EC: 26	21 peer-reviewed papers + 1 master thesis + 1 report from ENEA – Italian National Agency for New Technologies, Energy and Sustainable Economic Development + 1 BREF document + 2 websites	Lab tests	1 min – 12 weeks	All documents from Europe (mainly Italy, Belgium and the Netherlands) except for 6 papers (China, Taiwan, Canada, and U.S.A.)
<i>Construction materials</i>	Sand Lime Brick (SLB)	1	Report of the German Federal Association of Sand Lime Brick	Field test	Not reported	Germany
	Light-weight lime concrete – Autoclaved Aerated Concrete (AAC)	6	Peer-reviewed papers	Field tests	From 0 up to 33 years	5 papers: Japan 1 paper: Germany
	Pure air-lime mortars	1	1 literature review (21 publications)	The literature review considered field and lab tests	Different operating conditions tested	Valid for all contexts
	Mixed air-lime mortars	1	1 literature review (27 publications)	The literature review considered field and lab tests	Different operating conditions tested	Valid for all contexts
<i>Construction materials</i>	Hemp-lime	9	8 peer-reviewed papers; 1 meeting proceedings or seminars	5 documents: Life Cycle Assessment (LCA) 3 documents: lab test 1 document: lab test and LCA	Lab test: 28 days – 10 months	8 documents: Europe (France, United Kingdom, Italy, Latvia) 1 document: India
<i>Civil engineering</i>	Soil stabilisation	6	2 peer-reviewed papers; 4 meeting proceedings or seminars	2 documents: field tests 1 document: lab tests	Field tests: 3–34 years Lab tests: 3–90 days	Germany; U.S.A.; Belgium
	Asphalt pavements	0	7 documents but none reporting information about the carbonation rate of lime in asphalt	-	-	-
<i>Environmental protection</i>	Drinking water	2	2 peer-reviewed papers about water softening	Full-scale field tests	-	Valid for all contexts
	Wastewater/ Biosolids treatment	2	Peer-reviewed papers about biosolids	Field tests	Not reported	Germany
	Sludge/ Dredging sediments treatment	2	Peer-reviewed papers	Field tests	From 3 to 34 years	Germany and U.S.A.
	Flue gas purification systems	NC: 5	4 peer-reviewed papers + 1 BREF document	4 full-scale tests	Few seconds	Europe (mainly Italy and United Kingdom)
		EC: 18	14 peer reviewed papers; 1 website; 1 conference presentation; 1 patent document; 1 BREF document	11 documents related to lab tests 6 documents about a field test experience	Lab tests: 10 min–25 h Field tests: 20 min	16 documents related to Europe (mainly United Kingdom and Italy), 1 document related to China
	Acid mine drainage	2		Field tests	Not reported	Valid for all contexts

(Continued)

Table 2. Continued.

Lime market sector	Lime application	N° total sources	Type of source	Type of test (field vs. lab test)	Test duration	Geographical context
<i>Environmental protection Agriculture</i>	Soil neutralisation	0	1 company website; 1 peer-reviewed paper 12 documents consulted but none with useful information	-	-	-
	Increment of soil organic carbon	2	Peer-reviewed papers	Field tests	70–80 years	Germany and France
<i>Chemical industry</i>	<u>Calcium carbide</u>	0	6 documents but none reporting information about carbonation rate of carbide lime because it depends on its use	-	-	-
<i>Non-ferrous applications</i>	Base and precious metals, lithium	Many documents on the theme were consulted (Copper: 14 documents; Zinc-Lead: 22; Nickel: 14; Gold: 4; Platinum Group Metals: 4; Lithium: 13) but no relevant quantitative information was derived				
	Titanium dioxide production - Red gypsum	7	Peer-reviewed papers	Lab tests in conditions of enhanced carbonation	5 min - 3 h	6 papers: Malaysia 1 paper: Spain
	Aluminium production - Red mud	NC: 1 EC: 11	1 peer-reviewed paper 10 peer-reviewed papers 1 report from an industrial company	Field test 10 lab-tests; 1 experience of full-scale	Not reported 20' - 1 d	1 paper: Australia and China 1 paper: Europe (Greece) 10 documents: contexts outside Europe, i.e. Canada, India, U.S.A., and Korea Europe
<i>Pulp and paper – Precipitated Calcium Carbonate (PCC)</i>	Traditional ¹	1	BREF Document	Real experience in EU	-	Europe
	<u>Slag</u> ²	13	11 peer-reviewed papers; 2 university theses	12 documents: lab tests 1 document: field test	Different operating conditions tested	9 documents: Europe (Finland, Turkey, and Belgium) 4 documents: Brazil, Japan, Korea, and U.S.A.

¹Traditional = production via carbonation of hydrated lime; ² Slag = production via indirect carbonation of iron and steel slags.

NC = natural carbonation; EC = enhanced carbonation. The colour indicates the data robustness: applications with conclusive scientific data (bold and underlined), applications with less conclusive scientific data (only underlined) and no information available (no underlined).

From the analysed literature, the total content of Ca, expressed as CaO, is on average about 42.3% and that of Mg, expressed as MgO, is about 6.9%. The carbonation rate can reach up to about 30% in natural conditions and about 60% when the operating conditions are optimised, i.e. finer particle size and higher area in contact with CO₂ as shown in Table 4.

Among the other analysed sectors, a carbonation rate very close to 100% is reported for pure air-lime mortars (80–92%), for the conventional production of Precipitated Calcium Carbonate (PCC) in pulp and paper applications (85–93%) and for the softening of water in drinking water applications (100%). The complete carbonation of lime in drinking water applications was assumed by Scholz et al. [8] and confirmed by Wiley [9].

Regarding the construction materials sector, information about the carbonation rate of mortars is robust and mainly results from the literature reviews carried out by EESAC, who analysed the carbonation rate of two types of mortars: pure air-lime based mortars [10] and 'mixed' air-lime mortars, that are a

mixture of air-lime and hydraulic binder [11]. 'Air lime' mortars are made of hydrated lime and harden as a result of their exposure to atmospheric CO₂ and the formation calcium carbonate. Thus, carbonation is the hardening process of air lime mortars. Other types of mortar are a mix of lime and a strong hydraulic manufactured binder, e.g. Portland cement. In total, 190 publications provide data for carbonation for both types of mortars. After a pre-selection and check of the publications, 21 of them were considered reliable and adequate for air-lime mortars and 27 for mixed air-lime mortars. Pure air-lime mortars show a relatively high carbonation rate in the long-term, i.e. 80–92% after 100 years, while mixed air-lime mortars have a carbonation rate that is one-fourth of pure air-lime mortars (20–23%). The sector of Conclusive information is available hemp-lime construction materials due to the extensive work and publications carried out particularly in the United Kingdom and France, where this construction material is more widely used. The components of hemp-lime material are hemp shiv, i.e. the chopped woody core of the

Table 3. Literature-derived carbonation rate of lime in natural and enhanced conditions for each application.

Lime market sector	Lime application	Market share (%)	Natural carbonation rate (%)	Comments	Enhanced carbon. rate (%)	Comments
<i>Iron and steel industry</i>	Iron slags	41	Negligible even after 100 years	Ca and Mg are usually present in the form of silicates or in other low reactive compounds	7%-31%	Range derived only from lab tests. Minimum value for the direct route, maximum value for the indirect route
	Steel slags		5% (4 months) - 28% (\approx 1 year)	(1) 1 year is required for a good carbonation (2) huge piles of slag with low porosity should be avoided	39%-56% NOTE: \approx 45% in the PCC application ¹	Range derived only from lab tests. Values for direct route, slurry phase carbonation (most promising route)
<i>Construction materials</i>	<u>Sand Lime Brick (SLB)</u>	2.9	30% (age of samples not reported)	Value only from 1 document, from 25 years ago	-	-
	<u>Light-weight lime concrete - Autoclaved Aerated Concrete (AAC)</u>	4.4	30% (10 years) - 60% (30 years)	(1) The carbonation rate cannot be attributed all to lime due to the presence of CaO in the cement (2) Values from a Japanese research \rightarrow use in European contexts with similar climate conditions	-	-
	Pure air-lime mortars	0.4	80-92% after 100 years within 190 mm depth from the surface	Low uncertainty	-	-
	Mixed air-lime mortars	2.9	20-23% after 100 years within 190 mm depth	Low uncertainty (considering one fourth of the pure air-lime mortars value)	-	-
	Hemp-lime	0.1	55% after 91 days within 50 mm depth from the surface	Good consistency among the data found. Uncertainty given by the influence of hemp-lime composition on carbonation rate.	65%	Only one lab test study
<i>Civil engineering</i>	<u>Soil stabilisation</u>	6.4	37% after 34 years	Value from 1 field study valid for a lime dosage of 2.5% and German climate conditions	80%	Lab-test result on soil sample with 5% lime content after 3 days of curing
<i>Environmental protection</i>	Asphalt pavement	0.1	Not available	-	-	-
	Drinking water	2.1	100% presumably instantaneous	-	-	-
	<u>Wastewater/ Biosolids treatment</u>	4.3	Conditioning: 49% Post-stabilisation: 43%	Uncertainty due to: (1) values based only on 1 source, of 20-30 years ago (3) not reported the period of time between lime dosage and the analysis	-	-
	<u>Sludge/ Dredging sediments treatment</u>	2.9	35% after 30 years	High uncertainty \rightarrow analyses related to the same reclamation technology, but applied to soil	-	Enhanced carbonation has been recently tested but data only related to these as cement binder are available
	Flue gas cleaning systems	7.8	32% (lime reaction directly with the flue gas in a few seconds)	Value derived from 16 tests performed in European plants	Carbonation of Air Pollution Control Residues (APCR) 11-76% (lab tests) 27-34% (field tests)	The industrial experience is implemented in the UK by Carbon 8 Aggregates Ltd
<i>Agriculture</i>	Acid mine drainage	0.6	Negligible	Indications from full-scale plants	-	-
	Soil neutralisation	3.0	Not available	Focuses only on limestone applications	-	-

(Continued)

Table 3. Continued.

Lime market sector	Lime application	Market share (%)	Natural carbonation rate (%)	Comments	Enhanced carbon. rate (%)	Comments
<i>Chemical industry</i> <i>Non-ferrous applications</i>	Increment of soil organic carbon (SOC)		1–77 kgC/ha/year increment of SOC (after 70–80 years)	Uncertain range → It derives only from 2 studies		
	<u>Calcium carbide</u>	0.3	Depending on the actual use of the carbide lime, e.g. equal to PCC carbonation rate when carbide lime is used to manufacture PCC			
	Base and precious metals, lithium Titanium dioxide production – Red gypsum	1.5	The literature on the theme is almost absent. Based on the little collected information, the carbonation processes in solid residues (slags and/or tailings) should be modest No indication		Direct route: 10%–25% (first value for aqueous solution second value for the adding of NH ₄ OH) Indirect route: 60%	Uncertainty due to: - documents on lab tests - estimate based on the Ca content of the sample (not reported if it derives from lime or from limestone) - not considered the possible CO ₂ emission from limestone used in the TiO ₂ production process during the neutralisation
	<u>Aluminium production - Red mud</u>	1.4	11.5% (fresh sample, no indication on the exact age)	The CO ₂ sequestration is mainly due to its content of NaOH	11.5%	CO ₂ uptake comparable to that of natural ageing → The limiting factor is the Ca content rather than the operating conditions of the carbonation process
<i>Pulp and paper</i>	<u>Precipitated Calcium Carbonate (PCC)</u>	5.9	85–93%	Not 100% rate due to lime impurities and technical inefficiencies of the calcination and the carbonation process	-	-

¹Grosso et al. [26] made a specific review about the production of PCC via indirect carbonation of steel converter slag.

The colour indicates the data robustness: applications with conclusive scientific data (bold and underlined), applications with less conclusive scientific data (only underlined) and no information available (no underlined).

stalks of the hemp plant (*Cannabis sativa*), and air-lime binder with pozzolanic cementitious or hydraulic lime additives and in some cases, surfactants [12]. The air-lime binder is hydrated lime that, during the use phase of the hemp-lime construction material,

carbonates by reacting with atmospheric CO₂ and forming calcium carbonate [13]. The hemp-lime carbonation rate shows a good consistency around the value of 55% for hemp-lime with similar composition.

Table 4. Conditions for effective enhanced carbonation in conclusive applications.

Conclusive applications	Conditions for effective enhanced carbonation
Steel	Fine particle slags High contact with CO ₂
Pure air-lime and mixed mortars	Lower wall thickness Contact with CO ₂ Mortar composition (less cement higher carbonation rate)
Hemp-lime	Composition Contact with CO ₂
Drinking Water	Addition of coagulants and flocculating agents
Flue gas cleaning systems	CO ₂ concentration Lower SOx concentration Relative humidity Temperature Reaction time
Aluminium	Composition of red mud Contact with CO ₂
Pulp and paper	Contact with CO ₂ No impurities in lime

For Air Pollution Control Residues (APCR), lime is used for removing the acid compounds (HCl, SO_x, HF) contained in flue gases generated from combustion plants, in particular coal-fired power plants and waste incineration facilities. The flue gas purification process can be semi-dry or dry, which affects whether milk of lime or hydrated lime is used [14, 15]. In semi-dry processes, lime is supplied as aqueous solution or suspension, i.e. as milk of lime or as a lime slurry solution. During the reaction with the flue gas, the water evaporates and the reaction products are dry. In dry processes, powder hydrated lime is directly supplied as sorbent. For both processes the reaction products are separated from the flue gas in a conventional dedusting unit (typically a baghouse filter). During the flue gas purification, lime reacts with HCl, SO_x and HF but also with CO₂,

Table 5. Minimum and maximum natural carbonation rates of less conclusive applications and percentage of the process CO₂ emission reabsorbed through carbonation.

Application	Market share	Natural carbonation rate		Percentage of carbonation Max natural carbonation
		Min	Max	
Sand Lime Bricks	3%		30%	1%
Light-weight lime concrete	4%	30%	60%	3%
Soil Stabilisation	6%		37%	2%
Wastewater	4%	43%	49%	2%
Sludge Treatment	3%	43%	49%	1%
Calcium Carbide*	0.3%	-	95%	0.3%
Total	21%	-	-	10%

*The maximum carbonation rate of calcium carbide is assumed equal to the PCC one.

forming calcium carbonate. A carbonation rate in APCR of about 30% is reported, which can increase to about 60% when the operating conditions (CO₂ concentration, SO_x concentration, relative humidity, temperature, reaction time) are optimised as indicated in Table 4.

Lime in the form of quicklime or hydrated lime is used in the Bayer process, where bauxite ore is refined for alumina extraction. During the Bayer process, bauxite is digested in a caustic liquor of Na and Ca-hydroxides. This process generates two output streams: a liquor rich in alumina for subsequent aluminium production, and a solid residue, called red mud, which is disposal. The carbonation rate in aluminium production is low for both natural ageing and enhanced carbonation. In this case, most of the CO₂ sequestration is actually due to the use of sodium hydroxide rather than lime. The literature about lime carbonation in aluminium application is quite abundant and results are promising.; However, except for the APCR treatment, only lab tests have been performed so far. A strong push towards upscaling such processes to the industrial scale is highly encouraged.

Concerning PCC, no detailed information is available on the type of process and operating conditions applied in the PCC plants, the flows reported in the BREF [16] (600–660 kgCaO/tPCC) were taken as a reference to estimate the overall efficiency of the PCC production, which resulted in the range 85–93% (computed with respect to the stoichiometric lime consumption of 560 kgCaO/tPCC, Reactions 2 and 3).

The solid residues of both, the flue gas purification and the ferrous and aluminium production sectors can be used as a feedstock material for effective CO₂ sequestration during enhanced carbonation.

3.2. Lime applications with less conclusive scientific data on carbonation

Around 21% market share of lime applications according to Table 1 have less conclusive carbonation data, due to the lower number of relevant publications. The natural and enhanced carbonation rates of these applications are reported in Table 3.

For construction materials, not all the applications have very robust studies available. In particular, for light-weight lime concrete (also called aerated autoclaved concrete – AAC) a 60% carbonation potential was recorded after 30 years of service life under natural conditions [17, 18]. However, this rate cannot be attributed only to the lime, since some free CaO is also present in the cement used as a raw material. Regarding Silica Lime Brick (SLB), a 30% carbonation rate was reported during the service life (the age of the samples not being reported). However, this is based on one single literature source dating back 25 years [19]. New analyses on samples taken from European buildings with different ages in terms of service life should be performed to confirm the amount of sequestered CO₂ previously indicated and to better understand the time evolution of the carbonation process in outdoor ambient conditions.

A similar conclusion is drawn for the use of lime in the treatment of biosolids in wastewater treatment plants (for conditioning or sanitation after a dehydration unit). Two literature sources report a 40–50% carbonation rate for stocked biosolids, but the assessments were performed 25–30 years ago. Moreover, the time elapsed between the lime dosage and the sampling is unknown [20, 21]. When lime is used for the treatment of dredging sediments, the CO₂ permanently captured is 35% after 30 years [22]. Nevertheless, this rate is highly uncertain because it is related

to the same reclamation technology applied to soils and not to sediments.

Further research is needed to confirm the carbonation rate of soil stabilisation and in calcium carbide applications. Regarding the former, only one study provides a carbonation rate, while calcium carbide carbonation rate depends on the actual use of the carbide lime, which is the by-product of the main application of calcium carbide, i.e. acetylene production [23]. More information on the uses of calcium carbide manufactured from lime (i.e. how much of it is used for PCC, wastewater treatment, soil stabilisation and others) and on soil stabilisation carbonation is necessary.

In Table 5, the minimum and maximum carbonation rate for less conclusive applications are summarised. The market share of an application is calculated as the percentage of the calcination CO₂ emission for producing the lime used in that application with respect to the total calcination CO₂ emission for producing the lime used in all applications, given that 786 gCO₂ are emitted per kg of lime by stoichiometry. Then, the percentage of CO₂ reabsorbed through carbonation was calculated by multiplying the maximum natural carbonation rate by the market share of that application. The results are reported in the last column of Table 5. As a result, about half of the CO₂ emitted by the less conclusive applications is reabsorbed through the carbonation, which corresponds to 10% of the total emissions from lime calcination considering the maximum natural carbonation rate.

3.3. Lime applications with no available information on carbonation

Lime applications with no available information on carbonation accounted for 18% of European lime market sales in 2018. They are: agriculture, acid mine drainage, some sectors in the non-ferrous metallurgy without aluminium (production of base metals, of precious metals, and of lithium), asphalt pavements, soda and petrochemical manufacture. In all these applications the scientific literature on the topic is almost non-conclusive. In particular, for agriculture the existing literature focuses on the application of limestone and dolomite as liming materials, while for the treatment

of sediments indications are available for the reclamation technology (solidification and stabilisation) but relate to the soil not lime. In the case of non-ferrous metals, no information was derived apart from the overall amount of calcium in the slags and/or tailings and its mineralogical composition.

Other minor applications in construction materials, chemical industry and other industrial consumers were not assessed since no information about lime carbonation was identified in the literature.

3.4. Carbonation rate over time for conclusive applications

The carbonation rate over time depends on the specific lime application. Information about the carbonation time was found for all conclusive applications except for the aluminium industry. Lime used in flue gas purification, drinking water, pulp and paper will precipitate carbonate instantaneously, while lime applied in construction materials (mortars and hemp-lime) will carbonate during the whole lifetime of the building. From the assessed literature, the equations that link the carbonation rate to the lifetime of construction materials are the followings:

$$x = (K\sqrt{t}) \quad (1)$$

$$CR = MCR \cdot \frac{x}{depth} \quad (2)$$

Equation (1) describes the CO₂ diffusion across the application thickness through Fick's law [24] where K is the carbonation constant, t is the time expressed in days and $depth$ is the construction material thickness from its surface that carbonates after 100 years; x is the input of Equation (2) where CR is the carbonation rate; MCR is the maximum carbonation rate. For pure air-lime mortars, MCR and K are equal to 80–92% (see Table 3) and $1 \text{ mm}/\sqrt{\text{day}}$ respectively, while K of mixed air-lime mortars is $0.25 \text{ mm}/\sqrt{\text{day}}$ [10, 11]. In both cases, the depth is 191 mm [10, 11]. The carbonation rates in time of pure air-lime mortars and mixed air-lime mortars are reported in Figure 1 (top right and top left).

Regarding hemp-lime, Hirst et al. [25] found that the carbonation depth is proportional to the square root of time, like for mortars. Furthermore, the

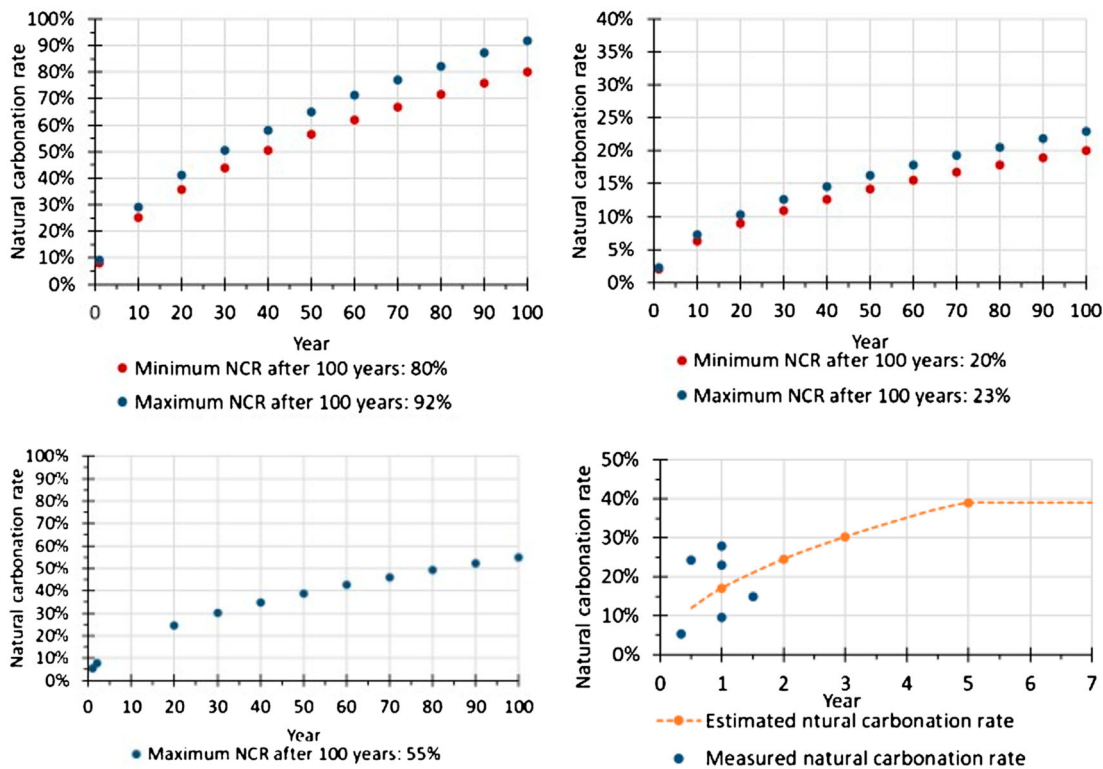


Figure 1. Natural Carbonation Rate (NCR) over time for pure air-lime mortars (top left), mixed air-lime mortars (top right), hemp-lime (bottom left) and steel slags (bottom right).

carbonation begins at an early age and it is lower at higher hemp-lime mix density, but after 91 days all cylindrical samples with diameter of 50 mm were fully carbonated [25]. As a result, *MCR* is 55% (see Table 3) while *K* is 5.24 mm/ $\sqrt{\text{day}}$ for a depth of 1001 mm and the carbonation rate in time is reported in Figure 1 (bottom right).

Figure 1 (bottom right) reports the carbonation rate in time for steel slags. Since no equation which links carbonation rate (*CR*) to time (*t*) was found in the literature, it was supposed as follows:

$$CR = a \cdot t^b \quad (3)$$

where *a* and *b* are parameters to be estimated. Considering the CO_2 uptake and relative age of the slag in Table 6 and 358 g CO_2 /kg slag as the maximum

uptake of steel slag found in the available literature Grosso et al. [26], *b* was estimated equal to around 0.5 and *a* equal to 0.009%/ $\sqrt{\text{day}}$. According to the equation, the minimum enhanced carbonation rate of 39% is reached after 5 years. The value is a little overestimated, considering that 2/3 of the free lime in steel slag will react within the first 5 years, whereas carbonation of calcium silicates and calcium aluminates will take more than 5 years, reaching a final carbonation degree of only 1/3. Thus, 39% can be considered as a theoretical maximum natural carbonation rate reached after 5 years, since the carbonated surface of the slags' pile is less porous and consequently the CO_2 diffusion in the pile will decrease over time. However, it has to be mentioned that typically the steel slags are stored in the open-air for 3–6 months only. While the equation based on Fick's law applied to mortars and hemp-lime is well known, the proposed equation for steel slags needs to be validated with further research. Thus, the natural carbonation rate of steel slags was considered equal to 5–28% (see Table 3) for the next evaluation in this subsection.

The percentage of the calcination emission reabsorbed through carbonation over 100 years for

Table 6. CO_2 uptake during natural ageing of steel slag.

Reference	Type of slag	CO_2 uptake (g CO_2 /kg slag)	Age of the slag
[34]	EAF (< 31.5 mm)	19.3	4 months
[35]	BOF (< 11 mm)	53	72 weeks
	EAF (< 11 mm)	34	48 weeks
	SS (< 11 mm)	100	48 weeks
[36]	BOF (< 16 mm)	82	1 year
[37]	BOF	87	6 months

Table 7. Percentage of carbonation rate over 100 years for applications with conclusive information obtained by multiplying market share and the maximum Natural Carbonation Rates (NCR).

Application	Market share	Maximum Natural Carbonation Rate (NCR)	Percentage of carbonation							
			T=0	T=1 year	T=5 years	T=10 years	T=20 years	T=30 years	T=50 years	T=100 years
Iron & Steel industry	41%	Maximum NCR: 28% after 1 year (Table 3) since open-air storage lasts 3–6 months	0%	11%	11%	11%	11%	11%	11%	11%
Pure air-lime mortars	0.4%	Maximum NCR: 92%; 100 years	0%	0.0%	0.1%	0.1%	0.2%	0.2%	0.3%	0.4%
Mixed air-lime mortars	2.9%	Maximum NCR: 92%; 100 years	0%	0.1%	0.1%	0.2%	0.3%	0.4%	0.5%	0.7%
Hemp-lime	0.1%	Maximum NCR: 55%; 100 years	0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.1%
Drinking Water	2.1%	Maximum NCR: 100%; instantly	2.1%	2.1%	2.1%	2.1%	2.1%	2.1%	2.1%	2.1%
Flue Gas Purification	7.8%	Maximum NCR: 32%; instantly	2.5%	2.5%	2.5%	2.5%	2.5%	2.5%	2.5%	2.5%
Aluminium	1.4%	Maximum NCR: 11.5%; assumed all at T=100 years as worst scenario since no information about time	0%	0%	0%	0%	0%	0%	0%	0.2%
Pulp & Paper	5.9%	Maximum NCR: 93%; instantly	5.5%	5.5%	5.5%	5.5%	5.5%	5.5%	5.5%	5.5%
Total	61%		10%	22%	22%	22%	22%	22%	22%	23%

conclusive applications, that account for 61% of the market share, was calculated (Table 7). As a result, considering the maximum natural carbonation rate (see Table 3), about 23% (after 100 years) of the total process emissions are reabsorbed through carbonation and with more than 95% of carbonation being already achieved after 1 year.

3.5. Carbonation as a carbon capture and storage process

Carbonation is under study as a carbon capture and storage process since the continuous growth of CO₂ atmospheric concentration and the ambitious objective of the Paris Agreement to limit global warming to well below 2°C require not only to achieve zero emissions as early as possible, but also to implement negative emissions [2], i.e. the removal of CO₂ from the atmosphere. The deployment of carbonation is under study in processes for capturing CO₂ from industrial flue gases [27] or directly from the atmosphere [28]. In these cases, the quicklime is obtained by calcining again the calcium carbonate generated during the capture phase and it is utilised again as sorbent, while the separated stream of CO₂ is sent to a permanent storage or it is used in industrial processes such as methanol synthesis. Alternatively, the quicklime is not produced again through calcination and the obtained calcium carbonate is used as a product, e.g. as a cement additive [29, 30]. Renforth [31] studied the potential of the different alkaline materials currently produced worldwide to achieve negative emissions

through the carbon dioxide removal process called Ocean Liming (OL), an enhanced weathering process.

In addition to these applications of lime carbonation with the specific aim to mitigate climate change, the carbonation of lime products is currently absorbing CO₂ from the atmosphere as analysed in the previous sub-chapters, but this is not taken into account in the National Greenhouse Gas Emission Inventories as a carbon sink, while the CO₂ calcination emissions when producing lime and cement are considered. To overcome this issue, Andersson et al. [32] introduced three calculation methodologies for evaluating the annual CO₂ uptake by cement products during their lifetime based on many studies in the literature following the Intergovernmental Panel on Climate Change (IPCC) guidelines for emission inventories. The objective of the study was to include CO₂ uptake by existing cement-based products in countries emission inventories that already take into account the calcination CO₂ emissions. The simplified calculation methodology 'Tier 1' proposed by Andersson et al. assessed the annual CO₂ uptake by the existing cement products as a percentage of the annual calcination CO₂ emission, estimated at about 20%. This means that each year, 20% of the calcination annual CO₂ emission is reabsorbed by existing cement-based products, given that new cement products absorb more CO₂ than old ones. Xi et al. [33] assessed also the percentage of calcination emission reabsorbed through carbonation. Based on experimental works and a literature review, they estimated the

carbonation potential of cement products to account for 17.6% of the global CO₂ calcination emission, which are then re-absorbed during the entire lifecycle of concrete. They assessed also that cement mortar reabsorbs 100% of calcination emission during its lifecycle. The higher carbon uptake is because mortar is applied in thin decorative layers to the exterior of building structures with larger areas exposed to the atmosphere.

4. Conclusions

Since the carbonation process of lime can play a crucial role for tackling climate change, it is necessary to quantitatively assess it both in terms of total long-term CO₂ absorption and of carbonation evolution over time according to the different applications where this can take place.

This work has assessed the carbonation rate in the multiple applications where lime is used. For each application, a review was performed to assess the potential carbonation rate of lime in terms of natural as well as enhanced carbonation. Based on the robustness of the information about carbonation found in the assessed literature, the applications were grouped in three clusters: conclusive, less conclusive or with no information available.

Considering the maximum natural carbonation rates, the eight conclusive applications, which account for 61% of the European lime market in 2018, will reabsorb potentially 23% of the total process CO₂ emitted during calcination, while the less conclusive ones, which represent 21% of the European lime market, will reabsorb potentially 10% of the total process CO₂ emitted during calcination. In total this leads to 33% of the calcination CO₂ emission being reabsorbed by conclusive and less conclusive applications (which account for a total of 82% of the European lime market).

Finally, information about carbonation over time was found for all conclusive applications except for the aluminium industry.

The findings of this literature review are a starting point for including the role of lime products through carbonation in the overall carbon accounting, and to promote further research in the improvement of the robustness of the results and in the enhancement and optimisation of such process.

Data availability assessment

The data supporting the findings of this study are available within the article and its supplementary materials.

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