

Current state of industrial heating and opportunities for decarbonization



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ABSTRACT

The IPCC recommends keeping the global average temperature increase well below 2 °C, if not below 1.5 °C, by 2100 to avoid the worst effects of climate change. This requires achieving carbon neutrality shortly after 2050. In the United States, industrial emissions represent 22% of greenhouse gas emissions and are particularly hard to decarbonize, because (1) the processes emit CO₂ as a byproduct of chemical reactions and (2) these industries require high-grade heat input. This study focuses on some of these industries, namely cement, lime, glass, and steelmaking. This work details the incumbent kiln and furnace technologies and explores the developing processes with examples of existing projects that aim to reduce carbon emissions, such as carbon capture and storage (CCS), fuel switching, and other technological changes. We provide tools to evaluate the most appropriate low-carbon solutions at existing facilities and on new-build infrastructure while taking into account the local context and resources.

This paper highlights two states within the U.S. with a high concentration of cement, lime, glass, and steel-making facilities, California and Pennsylvania. The emissions from cement, lime, and glass facilities in California total 8.5 MtCO₂eq/yr. About 6.3 MtCO₂/yr (7.1% of in-state industrial emissions) could be captured from cement and lime facilities, transported, and stored in sedimentary basins below the Central Valley. Replacing 20% of coal by biomass could also reduce the fossil emissions by 0.5 MtCO₂/yr (6.2% of in-state industrial emissions) without making changes to the facilities. In Pennsylvania, heavy industry (cement, lime, glass, and steelmaking) emits about 9.4 MtCO₂eq/yr. Most of the facilities are located near sedimentary basins, facilitating the development of CCS. In addition, the presence of low-carbon energy sources can help in the deployment of electrified processes. Also, industrial byproducts such as steel slag and fly ash can be reused in low-carbon concrete mix. As shown with these two examples, there are many strategies leading to the deep decarbonization of the economy and they need to be adapted to the local context.

1. Introduction

1.1. Global context for industrial decarbonization

The scientific community has linked the increase of anthropogenic greenhouse gas (GHG) release in the atmosphere to global climate change, which has been theorized since 1856 by Eunice Newton Foote [1]. In 2019 global greenhouse gas emissions reached 59.1 ± 5.9 GtCO₂eq/yr [2], primarily from the combustion of fossil fuels. CO₂ represents the majority of these emissions (76%). Methane (16%), nitrous oxide (6%), and fluorinated gasses (2%) are responsible for the

remaining quarter of the greenhouse gas effect [3]. The baseline scenario of the IPCC predicts a global temperature increase between +3.7 °C and +4.8 °C in 2100 compared to pre-industrial levels [4], which would have major consequences on human lives and biodiversity. In order to limit these negative effects, the IPCC recommends limiting the global average temperature increase to a maximum of 2 °C, if not below 1.5 °C [5]. This requires decarbonizing our economy and reaching carbon neutrality shortly after mid-century [5].

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1.2. U.S. contributions to global industrial greenhouse gas emissions

The United States emitted 6.7 GtCO₂eq in 2018, distributed between transportation (28%), electricity (27%), industry (22%), commercial and residential (12%), and agriculture (10%) [6]. Most greenhouse gas emissions in the transportation, electricity, commercial, and residential sectors are from the combustion of fossil fuels. Reducing the carbon footprint of these sectors can be addressed by replacing the energy source with renewable and low-carbon energy, which is a challenge that will not be covered here.

The industrial sector faces a separate challenge, as the processing of raw materials can generate CO₂ emissions as a by-product of chemical reactions. For example, in cement processing, the calcination of limestone to lime results in the production and emission of CO₂. These hard-to-avoid process emissions are the focus of this study. They require their own set of decarbonization solutions, in particular for the production of essential materials like cement, lime, glass, and steel, which will still be needed in a decarbonized economy.

In addition to process emissions, the industrial sector needs large energy inputs that generate stationary combustion emissions and indirect emissions. Stationary combustion emissions are those associated with the heating of raw materials through combustion of fossil fuels, biomass, or other wastes. These emissions are discussed further in this study, as the type of fuel combusted has a significant impact on the GHG emissions of the non-ferrous mineral (cement, lime, glass) and steel industries. Indirect emissions come from the purchase of electricity generated outside of the facility. These emissions were not considered in the study, as the facilities do not have control over the carbon intensity of the grid.

1.3. Production and emissions from the cement, lime, glass, and steel sectors in the U.S.

In the United States, direct process emissions from iron and steel production and metallurgical coke production reached 42.6 MtCO₂eq in 2018, and emissions from cement production totaled 40.3 MtCO₂eq the same year [6]. Lime and glass industries require high temperature inputs and face similar challenges to cement and steel industries and were thus included in this study. Direct emissions due to the production of lime and glass in the U.S. in 2018 were 13.2 MtCO₂eq and 1.3 MtCO₂eq, respectively [6].

These energy intensive processes are also large emitters of GHGs,

which need to be addressed to mitigate climate change. Fig. 1 shows the distribution of emissions of the mineral and steel industries in the U.S. for the year 2018, along with the amount of material produced and the CO₂ intensity of each product.

A large variety of decarbonization options are already on the market or in development today, including fuel switching, carbon capture and storage (CCS), and technological changes such as electrification or oxy-fired kilns. The choice of decarbonization pathway will depend on the availability of resources locally (raw materials, low-carbon fuels and energy, etc.), the readiness of the technology, and the cost. This study shows the spatial distribution and time of next likely update, along with local resources, to outline the best opportunities for each facility.

1.4. Kiln technologies and materials carbon intensity

The main piece of equipment of a cement, lime, glass, or steelmaking facility is a kiln or furnace, which reaches the high temperatures required to produce these materials. Kilns have different designs and heat sources based on the material produced and the demand for improvements within each individual process. Unique production requirements have driven the optimization of each type of kiln for its specific process. Rotary kilns, shaft kilns, hearth kilns, blast furnaces, basic oxygen furnaces, and electric arc furnaces are the main systems used to produce materials such as cement, lime, glass, and steel.

Across the globe cement and steel are major industries. Of which, rely on similar incumbent technology. The decarbonization pathways presented here are not limited to the context of the U.S., and can therefore, be applied in any region depending on the local resources, however how they are implemented may differ based on local policy. From a global perspective, a total of 4.10 Gt of cement and 1.86 Gt of steel were produced globally in 2019, emitting 2.3 GtCO₂ and 2.6 GtCO₂, respectively [10,11]. These emissions are distributed such that 1.8 GtCO₂ and 3.1 GtCO₂ are direct and process emissions, respectively [11]. These emissions are expected to rise to 2.3 GtCO₂/yr for the cement industry and 3.3 GtCO₂/yr for the steelmaking industry in 2050, whereas the 2 °C scenario from the IPCC advises to reduce the emissions to 1.7 GtCO₂/yr and 1.3 GtCO₂/yr by mid-century [12–14], respectively. Since 2000, the production of steel has approximately doubled while the total CO₂ emissions stayed roughly the same. This is due to the development of more efficient processes and the increased use of EAFs and of scrap steel [7,14]. The share of production from EAFs reached about 29% of the global production in 2018, and is predicted to rise,

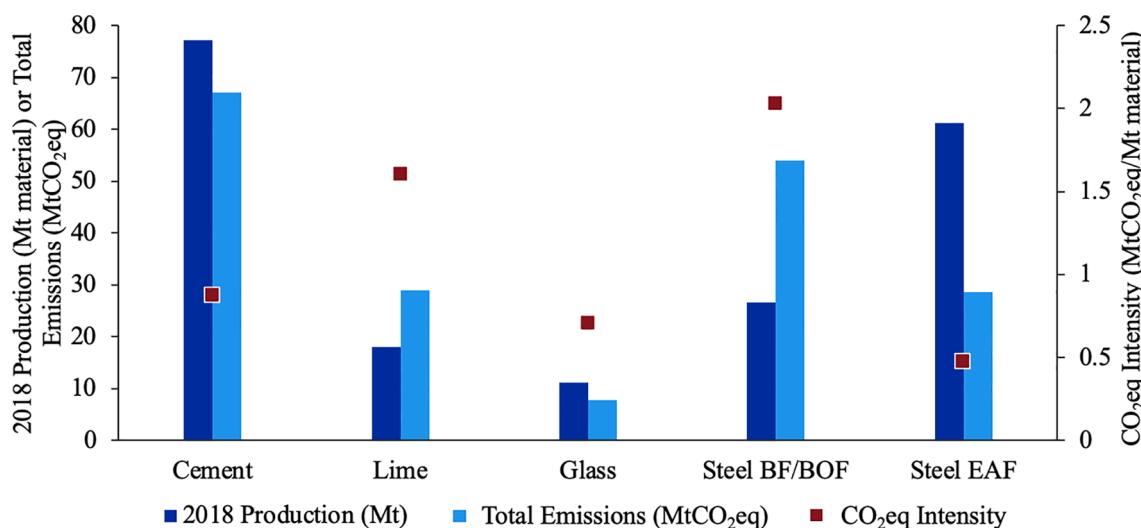


Fig. 1. Distribution of CO₂eq emissions from industrial processes in the U.S. in 2018 (BF/BOF: blast furnace / basic oxygen furnace, EAF: electric arc furnace) compared to the total production of the material. Data markers refer to the carbon intensity measured by the total emissions divided by the total production, resulting in an estimate of MtCO₂eq per Mt of material produced [7–9].

particularly in developing countries in the coming decades [14,15]. GHG emissions from the cement industry have roughly doubled since 2000, while the production of cement has nearly increased by a factor 2.5 [16], indicating only a slight decrease in carbon intensity. This is due to the lack of major technological changes to reduce carbon emissions in the cement industry, which would need to take place in order to decrease the emissions to be in line with the IPCC goals.

The selection of decarbonization options by a facility would depend on the local context. As noted previously, the kiln or furnace is a major piece of equipment at a facility. Replacing this unit will require a major investment by the company, which is most likely to happen if the facility's equipment is old, and the company has already invested in renovating it. On the other hand, investment in a carbon capture unit is more likely to happen at facilities where the equipment still has a long lifetime. To foresee likely changes, it is important to know the date of last renovation and the lifetime of current equipment.

1.5. Outline of the study

This study details the incumbent kiln and furnace technologies and explores different decarbonization options of the cement, lime, glass, and steel industries with the perspective of local availability of materials in the U.S. California and Pennsylvania are analyzed more deeply to outline opportunities in these two states.

2. Existing kiln and furnace technology

2.1. Non-metallic mineral industries

2.1.1. Rotary kilns for cement and lime production

Rotary kilns are primarily used in the cement and lime industries to produce clinker and lime, respectively. Fig. 2 shows a side view and cross-sectional view schematic of a rotary kiln. This paper focuses on the kiln technology specifically, and the complete cement production process is documented elsewhere [17]. For clinker production, feedstock made up primarily of limestone (CaCO_3) and clay, is typically fed into pre-calciner cyclones that raise its temperature to 600–900 °C in a preheating step. The pre-calciner cyclones are heated using exhaust gasses from the downstream pre-calciner and rotary kiln. The feedstock then enters the pre-calciner. In the pre-calciner, the feedstock is further heated. In these conditions, the limestone is calcined, releasing CO_2 , leaving lime (CaO), shown in Eq. (1). Just above half of the CO_2 emissions from this process originate from the calcination step [17]. The

pre-calciner and rotary kiln are often fired with coal, coke, or natural gas and can reach up to 1500 °C [8,18]. In the kiln, the limestone finishes calcination and the lime and clay fuse together, resulting in solid clinker. The solid clinker particles typically measure between 5 mm and 25 mm upon exiting the kiln, and rotation of the kiln ensures the clinker particles produced are uniform and nearly spherical [19]. Lime production is almost the same as clinker production, but the feedstock is only limestone and after it is calcined, the resulting product is lime (CaO). Additionally, the pre-calciner cyclones are often replaced with the vertical preheater, but it operates in the same capacity.



In the clinker and lime production processes, the calcination of feedstock materials (mainly limestone), and the combustion of fuel in the kiln results in CO_2 emissions. To reduce costs and potential emissions, some cement facilities use biomass or municipal solid waste (MSW) as fuel in place of fossil fuels. When MSW, such as tires, are used to fire cement kilns, it can reduce demand for other fossil fuels, such as coal or natural gas, but a life cycle assessment is needed to determine the total emissions savings. For example, nearly 44% of cement plants in the U.S. use tires or tire-derived fuel (TDF) to power their kilns [18]. Additionally, two cement plants, located in Texas and New York, use waste automotive carpet and one cement plant in California uses pistachio shells to supplement kiln fuel [18]. In the lime industry, fuel switching has not gained much traction, and all lime rotary kilns use forms of fossil fuels for heating to avoid a reduced-quality lime product [21]. The current distribution of fuels used in rotary kilns for cement and lime production in the U.S. can be seen in Table 1.

2.1.2. Shaft kilns for lime production

Nearly 40% of lime producers use shaft kilns rather than rotary kilns [22]. When a shaft kiln is used, the limestone is preheated in a vertical preheater before entering the kiln. The lime enters the shaft kiln at the top and moves downwards through it. There are burners nearly midway down the shaft where fuel is burned to heat the limestone to calcination temperatures (above 800 °C). Air enters through the bottom to assist in keeping the lime moving through the shaft and to allow the lime to cool before exiting the kiln at the bottom. The CO_2 that exits the kiln is both from the calcination of lime and from the fossil fuels used in the burner.

The fuel used in lime kilns is similar to that of cement; however, there seems to be less adoption of fuel switching. Roughly 26% of lime shaft kilns currently use biomass as fuel [18]. When biomass is used in lime kilns, it is often done so in the side burners of the shaft kiln. This method has proven to keep NO_x emissions down and maintain the quality of the lime product [21]. Table 2 shows the distribution of fuels used in the shaft kiln lime production and the associated emissions in the U.S. lime industry.

2.1.3. Hearth furnaces for glass production

Glass making usually takes place in a hearth furnace, as shown in

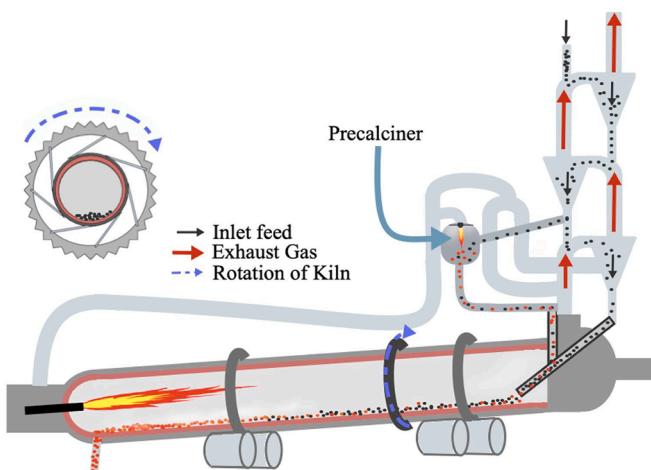


Fig. 2. Side and cross-sectional view of a rotary kiln, illustrating the feedstock entering through the pre-calciner cyclones and then entering the rotating kiln body. As the feedstock moves down the rotary kiln body, it increases in temperature, as shown by the color change, before exiting [20].

Table 1

Predominant fuel use in rotary kilns and emissions (MtCO₂eq) distribution in the cement and lime industries in the U.S. [18].

Dominant Fossil Fuel (s)	Number of Cement Facilities	Number of Lime Facilities	Total Emissions from Cement (MtCO ₂ eq)	Total Emissions from Lime (MtCO ₂ eq)
Coal	21	12	18.96	4.64
Natural Gas	7	5	5.83	2.57
Coal and Natural Gas	48	29	35.15	16.11
Other Fuels ^a	1	0	0.36	0
Unknown	14	0	6.71	0

^a Biomass and municipal solid waste (MSW) are not included in this category, as neither make up the predominant fuel used at cement facilities in the U.S.

Table 2

Fuel use and emissions (MtCO_2eq) distribution for shaft kilns in the lime industry in the U.S. [18,22].

Dominant Fossil Fuel (s)	Number of Shaft Kiln Facilities	Total Emissions (MtCO_2eq)
Coal	2	0.72
Natural Gas	4	1.23
Coal and Natural Gas	17	2.83

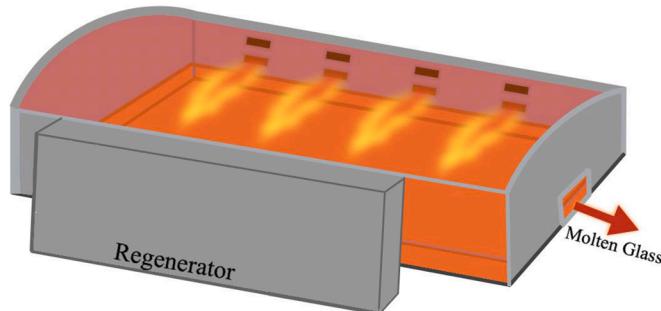
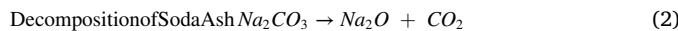


Fig. 3. Inside of a hearth furnace which heats glass feedstock until it is molten. The oxygen-enhanced atmospheric gasses are circulated through the regenerator and the molten glass exits, where it is later cooled to the desired temperature with the air circulated by the regenerator [22,23].

Fig. 3, heated using natural gas or oil-derived fuels and reaching temperatures between 1400°C and 2000°C . Hearth kilns are fed with silicate sand, soda ash, limestone, and recycled glass pieces. These are heated in an oxygen-enhanced atmosphere until molten in the hearth kiln. This molten material is tapped so it pools onto a bed of molten tin. The melting point of tin is substantially lower than the temperature of the molten glass, allowing the glass to cool and solidify, while floating on the top of the molten tin.

Some of the CO_2 emissions associated with this process are from the soda ash and limestone being calcined in the hearth kiln Eqs. (1)–(2)). Approximately 37% of the emissions from glass production are process emissions, while the other 63% are from stationary combustion [23]. In addition to the process emissions from glass production, the CO_2 emissions associated with operating the kiln are produced by the combustion of natural gas or oil-derived fuels used to produce high temperatures.



2.2. Iron and steel industry

There are two main processes used to produce iron and steel. One includes a blast furnace and basic oxygen furnace (BF/BOF) configuration while the other utilizes an electric arc furnace (EAF). The BF/BOF process is most often used in primary steelmaking, whereas the EAF is most often used for secondary steelmaking using recycled steel scrap. Here, we will only be discussing the furnace configuration of each production route, but full details of each process can be found in Bains et al. (2017) [17].

2.2.1. Integrated steel mill (BF/BOF process)

Steel mills that include sintering, coke production, blast furnace, and basic oxygen furnace processes, are referred to as integrated steel mills. Before entering the blast furnace, iron ore is ground down into pellets and heated with coke and lime to produce sinter. As shown in **Fig. 4**, sinter and limestone flux are put into the top of the blast furnace where they are met by hot blasts of air that come from the bottom. Inside the furnace, the raw materials reach white hot temperatures (2200°C) to form pools of liquid slag and molten iron. The slag produced at this step is made up of the lime and iron ore impurities. The molten iron is

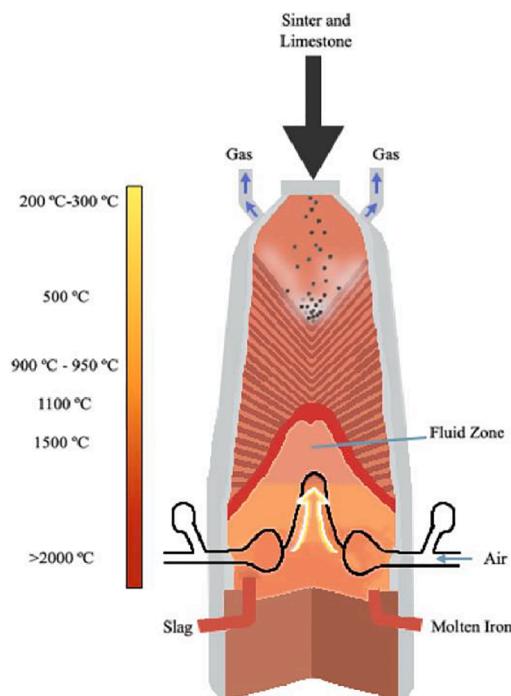
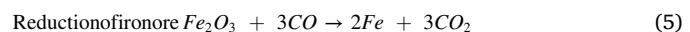
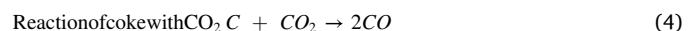
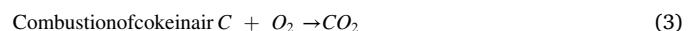


Fig. 4. Inside of a blast furnace showing sinter and limestone feedstock entering at the top, with air entering near the bottom and the outlet of steel slag and molten iron [24].

referred to as pig iron, which is known and utilized for its high carbon content.

The BF is heated using natural gas and coke. The natural gas is used to heat the air blown into the main body from below. As this hot air comes in contact with the coke, which enters from the top, the coke combusts. Together these create the 2200°C environment which reduces (removes the oxygen from) and melts the iron ore. The primary reactions that take place in the blast furnace are as follows: combustion of coke in air (Eqn. (3)), calcination of limestone (Eqn. (1)), reaction of coke with CO_2 (Eqn. (4)), and reduction of the iron ore (Eqn. 5). The process emissions that originate from the limestone calcination is approximated to make up 7% of the total emissions from the process. The process emissions from the reduction of iron are difficult to estimate because the coal used in the process serves a dual purpose; of providing heat as well as establishing a reduction environment of CO. Part of the emissions from the use of coal should be allocated to process emissions, while the remaining portion would be accounted for in stationary combustion emissions.



The pig iron that exits the blast furnace still contains carbon, sulfur, phosphorus, manganese, and silicon, some of which need to be removed to produce steel. Most of these impurities can be removed through the processing in a basic oxygen furnace (BOF), as shown in **Fig. 5**. Pig iron from the BF and scrap steel are fed into the BOF for processing. Once in the BOF, a lance is lowered to blow high purity oxygen at supersonic speeds over the scrap and molten iron. This causes the impurities to oxidize and produces heat. The temperature in the BOF is controlled by altering the amount of scrap steel and iron ore that is used in this step of the process. Steel is removed from the BOF and further additions can be made for alloying. Slag produced in this step is recovered and recycled.

In addition to the BF and BOF, some integrated steel mills also have

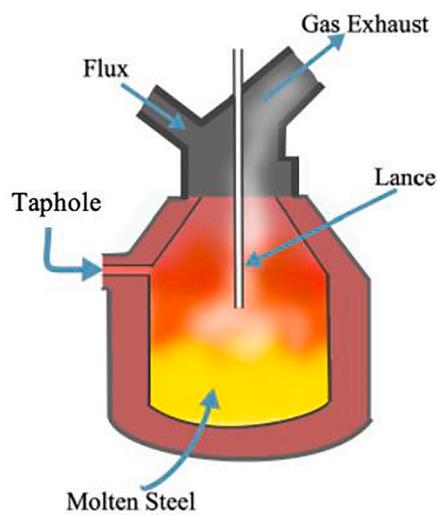


Fig. 5. Basic oxygen furnace (BOF) showing the central lance providing high-purity oxygen at high velocities, and a pool of refined molten steel at the bottom of the furnace. When the molten steel has reached the desired purity, the furnace is rotated and the molten steel flows out of the taphole [25].

on-site power plants, or “captive” power plants. These power plants ensure consistent provision of electricity and can often provide it at lower cost than if the plant was to rely on the grid. In addition to decarbonizing their BF/BOF process, these facilities also have the opportunity to generate electricity with a lower carbon intensity than the grid.

2.2.2. Mini mill (EAF process)

The other main method of producing steel is through the EAF. Steel mills with primarily EAF steel production are also referred to as mini-mills. The name comes from the fact that EAFs can be operated

efficiently on a smaller scale than a BF/BOF configuration. The EAF process is most often used in steel recycling but can be used for primary steel production when coupled with a direct reduction pathway. Direct reduction of iron (DRI) is used in place of the BF. Rather than melting the iron to remove impurities, the iron is heated to temperatures below its melting point in an environment of reduction gasses. These gasses drive off the oxygen contained in iron ore, producing metallic iron. Unlike pig iron (the product of a BF), the result of the DRI process, sponge iron, which still retains silica that must be removed in the EAF [26]. A combination of recycled scrap, sponge iron, and hot-briquetted iron (compacted form of DRI) is first loaded into an EAF chamber. The chamber is then shut, and graphite electrodes are lowered into the main body and treated with a strong electric current. This current produces an arc that heats and melts the iron, creating a molten pool. A cross section of an EAF can be seen in Fig. 6. Lime, feldspar, carbon, and oxygen are all used to react with impurities that create a slag pool which can then be separated and removed from the molten refined steel.

The reduction gas environment required for the DRI process is typically carbon monoxide (CO), hydrogen (H₂), or both (syngas). If coal is used, then CO is the reducing gas, and if natural gas is used, then the reducing gas is syngas. To achieve a CO or syngas environment, the heating of coal or a mixture of oxygen (O₂) and natural gas (CH₄) is typically used, resulting in direct CO₂ emissions. Although a pure H₂ environment does not directly result in CO₂ emissions, there may be CO₂ emissions associated with the process to produce the H₂ being used. The EAF process does not use fossil fuels directly, but depending on the fuel used for electricity generation, there are associated CO₂ emissions, which are expected to decrease in the coming decades, as the electricity grid decarbonizes [31]. There are also small amounts of process emissions to be considered with the EAF process. As the carbon from the iron is released, it binds with the oxygen being blown in to create carbon monoxide. This carbon monoxide partially burns in the oxygen-rich atmosphere and produces CO₂. Due to the high electrical current needed for EAF operation, the graphite electrodes used in this process also degrade at a rate of 1 to 2 kg/tonne of steel produced [32], resulting

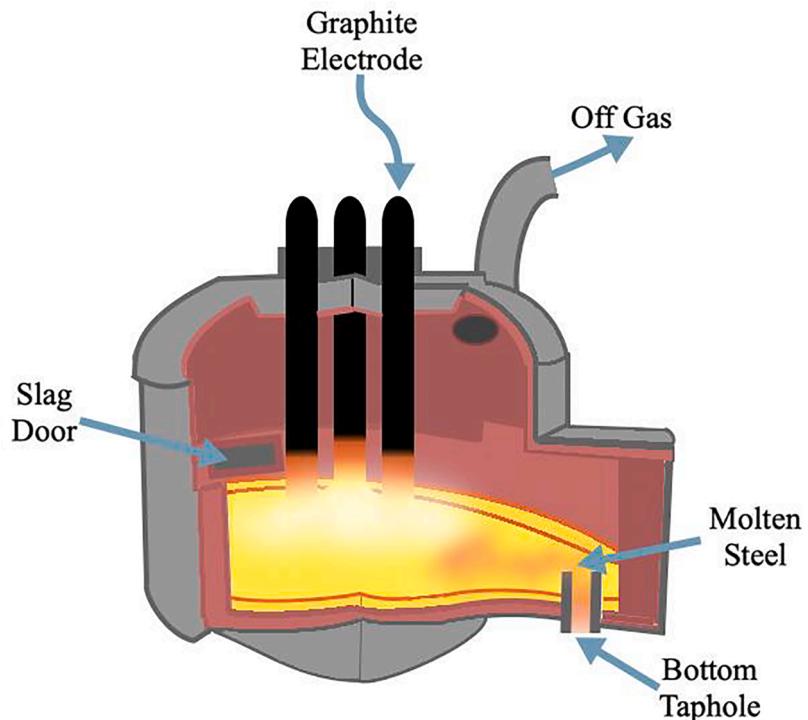


Fig. 6. Inside of an electric arc furnace (EAF) showing the graphite electrodes that conduct an arc with the steel feedstock to drive off impurities. The slag collects on top of the molten steel and is released through the slag door while the molten steel is collected through the bottom taphole [27–30].

in additional CO₂ emissions.

3. Innovative pathways to reduce CO₂ from industrial furnaces and kilns

Improvements can be made to the existing kiln and furnace technologies in these sectors to reduce the amount of CO₂ emissions that end up in the atmosphere. Although many existing and adopted technologies result in far fewer CO₂ emissions than those currently in use, favorable economics and production capacity of existing furnaces leads to continued production using furnaces with greater emissions intensities. For example, steel produced from scrap in an EAF results in markedly fewer CO₂ emissions than the BF/BOF route, but replacing existing BF/BOF production capacity, especially for higher quality steel products, is currently economically prohibitive.

3.1. Timescale for innovation

Industrial kilns and furnaces are designed to have long lifetimes because it is often expensive and time intensive to upgrade or replace them. Most kilns and furnaces have a steel outer shell and oxide bricks lining the inside. The oxide bricks are called refractory bricks and protect the metal outer shell from overheating and prevent deformation during operation. Due to the consumable nature of refractory bricks, they must be replaced on a regular basis to ensure proper operation of the kiln or furnace.

Table 3 summarizes the lifetimes of kilns and furnaces used in cement, lime, glass, and iron and steel production. These lifetimes reflect the time between substantial repairs, replacements, and needed maintenance. This time frame was specified because these activities often result in industrial downtime and can require financial investment on the order of adopting new kiln technology. Due to the capital investment required for an industrial kiln or furnace, retrofits and upgrades are more attractive when they coincide with a needed replacement. Given the longer lifetimes of these industrial kilns and furnaces, the timescale for retrofits and upgrades can range anywhere between 8 and 100 years. Estimates for the retirement year of rotary kilns, BF, BOF, and EAF were calculated for the facilities located in the U.S. using lifetimes of 40 years, 15 years, 100 years, and 65 years, respectively.

For each of the configurations, knowing the lifetime of the infrastructure helps to understand the dynamics of its replacement. This information coupled with the date of the last update of each unit indicates when a facility is planning for large investments and might be interested in changing for an upgraded or newer technology.

3.2. Energy and materials savings

3.2.1. Energy and materials savings for non-metallic minerals

There are few opportunities to reduce emissions from cement, lime,

Table 3

Lifetimes for kilns and furnaces used in cement, lime, glass, and iron and steel production.

Kiln or Furnace Type	Lifetime	Additional Service or Maintenance	Reference
Rotary Kiln for Cement and Lime Production	40–80 yrs	Refractory brick replacement every 1–3 yrs	[33,34]
Blast Furnace (BF)	15–20 yrs	–	[35–38]
Basic Oxygen Furnace (BOF)	100 yrs	Oxygen lance replacement needed after 400 heats	[39]
Electric Arc Furnace (EAF)	65 yrs	Graphite electrode replacement needed every 8–10 hrs of operation	[39,40]
Hearth Furnace	8–20 yrs	Refractory materials lifespan 14–18 yrs	[41–43]

and glass industries without changing the process equipment or deploying carbon capture and storage (CCS). The methods covered here relate to materials recycling and energy savings that can be achieved with existing process technology. Lime cannot be recycled because it is a product of a chemical reaction and faces energetic barriers to return to its original state. However, lime waste can be spread across soil to enhance nutrients [44].

Recycling cement and glass have proven to be promising in reducing CO₂ emissions through the lessened demand for virgin material. When concrete is decommissioned, hydrated and unhydrated cement can be separated from each other and from other constituent materials by use of smart crushing technologies [45]. The hydrated cement must be recycled by being heated in a cement kiln to drive off the water content [12]. In addition to reducing the need for raw material mining, this recycling process only results in CO₂ emissions from the fuel used in the kiln. The unhydrated cement can be re-crushed and reused in concrete [12]. The remainder of the concrete constituents can be reclaimed and recycled as an aggregate additive to road-based construction [12].

Another option for reducing CO₂ emissions from the cement industry is to decrease the demand for cement clinker by changing the chemistry of concrete to be less reliant on CO₂-intensive feedstocks. Clinker is the primary reactive component of ordinary Portland cement, which combines with water to bind together aggregates to produce concrete, while gypsum controls the reaction time [46,47]. Concretes that use blast furnace slag and fly ash are considered industry standard today, but these feedstocks could become less available as deep decarbonization of the industrial and energy sectors continue [47,48]. Some alternatives to replace these industrial waste materials have been identified as bauxite mine tailings, more commonly referred to as and natural pozzolan materials [48–52].

An important emerging substitute for cementitious material in concrete is limestone calcined clay (LC³), developed by Dr. Karen Scrivener. This concrete recipe consists of cement feedstock that utilizes both clay and limestone and results in a lower carbon footprint by up to 40% [53, 54]. Trial production of this cement was undertaken by the Siguaney Cement Factory in Cuba and JK Lakshmi in India [55–57]. In these trials, the cement produced can be used in a one-for-one capacity with most standard uses. Since its inception, LC³ has gained traction in the global cement industry and nearly 40 cement companies in 25 countries are interested in adopting it [54].

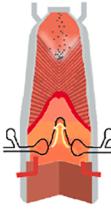
Glass can be recycled with no loss in product purity. Reclaimed glass with the purpose of recycling is called cullet. Increasing the cullet used in glass processing can reduce energy use by up to 3% and the need for primary glass feedstocks that contain carbon dioxide, such as limestone and soda ash [58,59]. The European Container Glass Federation (FEVE) estimates that when 100% cullet is melted to produce molten glass, CO₂ emissions are reduced by nearly 58% [59].

3.2.2. Energy and materials savings in the iron and steel industry

There are many options for reducing emissions in the steelmaking process, from retrofitting existing equipment to replacing infrastructure with innovative technologies. Energy efficiency and carbon emission reduction measures can be implemented with existing equipment in integrated mills and mini-mills, and these can offer low-cost and effective emissions mitigation options. **Fig. 7** outlines available energy efficiency and emissions reduction retrofits and new build opportunities for blast furnaces, basic oxygen furnaces, and electric arc furnaces.

3.2.3. Waste heat utilization

The cement and steel processes are energy intensive and produce waste heat that can be used by decarbonization processes. It is estimated that the technical potential for waste heat to power with a steam Rankine cycle to be around 2.1 GW at primary metal facilities and 0.7 GW at mineral plants in the U.S. at facilities producing over 5 MW of waste heat [60,61]. The lower bound for assessing the potential of the waste heat to power process is 232 °C, as commercially available



Blast Furnace (BF)									
Technology	Pulverized coal injection replaces coke	Natural gas injection replacing pulverized coal or coke	Coke oven gas (COG) and BOF gas injection	Recovery of BF gas	Hot blast stove automation	Heat recovery from granulating produced slag			
Energy and Materials Savings	3760 MJ/tonne coal injected	900 MJ/tonne of hot metal	0.9 tonne of COG or BOF gas replaces 0.89 tonnes of coke	65 MJ/tonne of hot metal	360 MJ/tonne hot metal	350 MJ/tonne pig iron			
Energy Savings (%)	17%	4.1%	0.03%	0.29%	1.7%	1.6%			
Basic Oxygen Furnace (BOF)									
Technology	Heat recovery from BOF gas		Improved oxygen management system	Heat loss reduction in ladle operation					
Energy and Materials Savings	920 MJ/tonne of steel		1.5% of electricity used for oxygen production	60% increase in heating efficiency					
Energy Savings (%)	4.2%		N/A	N/A					
Electric Arc Furnace (EAF)									
Technology	Ultra-high-power transformers	Bottom Gas Injection	Foamy slag	Pure oxygen injection	Adjustable speed drives for fans	Direct current (DC) for EAFs	Scrap preheating	Air-tight sealing	Side-by-side EAFs
Energy and Materials Savings	61 MJ/tonne steel produced	79 MJ/tonne steel produced	29 MJ/tonne steel produced	144 MJ/tonne steel produced	61 MJ/tonne hot metal	324 MJ/tonne steel produced	198 MJ/tonne hot metal	396 MJ/tonne hot metal	68 MJ/tonne hot metal
Energy Savings (%)	1.1%	1.4%	0.50%	2.5%	1.1%	5.6%	3.4%	6.8%	1.2%

Fig. 7. Blast furnace, basic oxygen furnace, and electric arc furnace energy efficiency opportunities, where “N/A” indicates that the data lacked granularity to calculate a percentage of energy savings. Notes: Baseline energy consumption for the above processes are 22.1 GJ/tonne steel for integrated steel mill operations, 5.8 GJ/tonne steel for EAF, and 2.9 GJ/tonne coke [32]. Electric arc furnace steelmaking consumes 350 to 600 kWh/tonne of steel produced, depending on design details [32].

technologies predominantly requires medium- to high-temperature waste heat sources [60]. Instead of being used for power generation, this waste heat could be used partially or in totality by a point source carbon capture unit, which requires temperatures around 100 °C [62].

Readily available waste heat would increase the efficiency of the carbon capture process as no additional energy supply would need to be built for this process. Pilorgé et al. [63] estimate that this industrial waste heat available on site could supply the energy to capture the totality of the 19.4 MtCO₂ of capture potential at iron and steel plants, and 45.9 MtCO₂ from cement kilns (about 80% of the capture potential). However, part of this heat might already be used at the facilities for processes requiring lower grade heat, as in the pre-calciner cyclones in the rotary kilns, and the reuse of the BF gas downstream in the steel production process, reducing their overall fuel inputs [32,64].

3.3. Retrofitting technologies: carbon capture and storage (CCS)

3.3.1. Readiness and projects

Carbon capture and storage includes three major steps: capture, transport, and storage. Transportation and storage techniques are already deployed at a commercial scale, mostly for the use of CO₂ in enhanced oil recovery (EOR) operations. This existing infrastructure could be used for transporting and storing industrially sourced CO₂ [65, 66]; however, its deployment is very sparse, and the private ownership of currently installed pipelines might be an obstacle for usage and connecting subsidiary pipelines. Today, most of this infrastructure is located around the Permian Basin, so would need to be expanded to service areas where there is more industrial activity, such as the Great Lakes region. New CO₂ pipelines and storage sites need to be built in the coming years in order to connect large industrial facilities to CO₂ injection sites and successfully deploy CCS.

Technologies for capturing CO₂ from flue gas are already in place at a few industrial facilities (e.g., Air Products' Port Arthur hydrogen production facility and the Archer Daniels Midland ethanol plant) [67], but

CO₂ capture requires technical adaptation to accommodate the differences in composition of flue gas from cement, lime, glass, or steelmaking facilities [68]. Among the cement, lime, glass, and steel industries, only one large scale CCS project is operational on a DRI unit of an iron and steel plant: the Abu Dhabi CCS project in the United Arab Emirates since 2016 [67]. The technology has proven effective and could be replicated at other DRI plants. DRI plants are not widely deployed in the U.S. yet but are expected to be looked at as viable replacements for BFs.

A partnership between Svante, LafargeHolcim, and Oxy Low Carbon Ventures is setting out to determine the feasibility of carbon capture at a cement plant in Colorado (U.S.), the LH CO2MENT Project, that is partly funded by the DOE [67,69]. The capture plant alone is estimated to cost around \$300 M [68]. This project is aligned with LafargeHolcim's commitment to reduce the carbon emissions of their cementitious materials to be 475 kg CO₂/tonne by 2030 [70]. Additionally, HeidelbergCement has approved a project in Brevik, Norway at the Norcern cement plant to implement CCS. The CCS system is estimated to capture 400,000 tonnes of CO₂ per year starting in 2024 [71]. This will be the first full-scale CCS cement plant in the world.

3.3.2. Capture potential

Implementing CCS generally does not require modifications to the industrial process in itself [12], which is attractive for many facilities. Using an air environment for combusting fuel in kilns and furnaces leads to a mix of CO₂, N₂, NO_x, and H₂O gas in the exhaust stream(s); the capture of CO₂ requires the separation of CO₂ from these other gasses. Some of these other gasses would have to be scrubbed ahead of the separation of CO₂, resulting in cleaner air for communities living in the vicinity of the facility [72].

Most studies consider that CO₂ capture can be achieved with a 90% capture efficiency [12,13,65]. A recent study questions this number and shows that 98% efficiency can be achieved with a marginal cost increase at industrial facilities [73]. Most of industrial exhaust concentrations are in the range 15–30% CO₂ [17,65,74,75]. For that range of CO₂

concentration, the total annual cost increases from \$56–60/tCO₂ for 90% capture to \$58–66/tCO₂ for 98% capture [73]. The remaining emissions need to be captured with direct air capture to reach net carbon neutrality. The marginal cost increase of less than \$10/tCO₂ is small compared to the cost of direct air capture, showing that achieving a 98% capture efficiency would decrease the overall cost of carbon neutrality for these industrial processes.

For practical and economic reasons, a facility is likely to implement carbon capture on the exhaust stream with the highest volume and concentration of CO₂ [65]. Also, CCS is currently the only solution to mitigate process emissions. Thus, there should be an emphasis made on implementing CCS to abate process emissions, as emissions from stationary combustion sources could oftentimes be solved by fuel switching or electrification. A common source of process emissions in the cement, lime, and steel industries is the use of limestone in the process. These process emissions can only be avoided through CCS [36].

In the cement and lime industries, the heating of reactants and burning of fuel both occur in the kiln, which reduces the exhaust to a single stack embodying all the process emissions from limestone calcination and emissions from fuel combustion. Point source capture thus had the potential to capture about 90% of the emissions from the cement and lime industries and avoid the release of roughly 57.8 MtCO₂ and 14.6 MtCO₂ in the atmosphere in 2017 [65,76], respectively.

At iron and steel facilities, there are multiple exhaust streams, with roughly 2/3 of non-process emissions originating from stationary combustion units. The major process stream is the blast furnace gas, which represents 78% of the process emissions [17]. Pilorgé et al. [65] estimate that implementing point source capture on the blast furnace exhaust could have avoided the emission of 19.4 MtCO₂ (70.2% of point source emissions from integrated steel mills) from iron and steel plants in 2017.

3.3.3. Storage and pipelines

To impact climate change mitigation, the captured CO₂ then needs to be stored safely and durably. Various options exist for the injection and storage of CO₂ in subsurface reservoirs [77–81]. Injection of CO₂ in sedimentary formations is the most established technique and has been demonstrated at a commercial scale by the Sleipner project since 1996 [82–84], the global capacity of sedimentary formations is evaluated in the range 8,200–34,700 GtCO₂ [77–81]. Alternatively, injection of CO₂ in basalt formations is promising with the success of the CarbFix (Iceland) and Wallula (Washington state, USA) pilot projects [85–87]. These projects have demonstrated the mineralization of CO₂ in basaltic reservoirs in less than two years [88,89], minimizing the risks of leakage. The capacity of on-land basalts is estimated to be in the 1000 s to 100,000 s GtCO₂ with similar additional capacity in submarine basalt formations [77,78,81]. Ultramafic rocks are also attracting interest, as they react more readily with CO₂ than basalt [77], and their storage capacity could be even greater from 60,000 to 600,000 GtCO₂ [77,78,81]. Local studies will have to be conducted to assess the true storage capacity of a specific rock formation. The action of secondary trapping mechanisms in geologic formations should mitigate any leakage from the reservoir and reduce the risk of leakage over time [88,90,91]. Well-regulated storage sites should retain over 98% of CO₂ over 10,000 years, down to 78% over the same period of time in a worst case scenario [92]. Concerns have also been raised about the feasibility of a fast deployment of permanent storage at the scale of Gt/yr, as this industry is facing cultural, legal, regulatory, and economic barriers [90].

For small volumes of CO₂ captured, transportation by truck is still viable, but for larger volumes and long-distance transportation to storage sites, pipelines are more economical [65]. Large-scale deployment of carbon capture with utilization and/or storage (CCUS) would thus require the development of sequestration sites and utilization facilities, as well as a CO₂ pipeline network. Trunk pipelines collecting CO₂ from multiple sources can benefit from economies of scale, while feeder pipelines serving individual plants do not. A CO₂ pipeline network already exists locally in the U.S. in the Permian Basin and along the Gulf

Coast for transporting CO₂ to EOR sites, for which 80% of the CO₂ is mined from natural underground CO₂ reservoirs [93]. The CO₂ pipeline network could be extended to include industrial sources of CO₂ and injection sites in saline aquifers. Studies have been conducted in the Midwest to link ethanol plants to sedimentary basins with EOR operations [94,95]. A pipeline collecting 4.30 MtCO₂/yr from 15 bioethanol plants would cost \$1.06 B in CAPEX and \$53 M in OPEX, while a more ambitious pipeline network involving 34 bioethanol production facilities and collecting 9.85 MtCO₂/yr would have a CAPEX of \$2.67 B, and an OPEX of \$131 M [94]. Sanchez et al. estimate a maximum levelized cost of pipeline transportation from bioethanol plants to geologic storage of \$19/tCO₂ and discuss the impact of credits [95]. Stolaroff et al. indicates that the cost of 40 km-long pipelines is roughly \$0.05–0.08/tCO₂/km and goes down to \$0.02–0.03/tCO₂/km for 1000 km-long pipelines based on a NETL cost model aggregating models of three research groups [96–100]. Similar work could be done to link other industrial facilities to storage facilities. However, as cement facilities are more geographically distributed from each other than blast furnaces (mostly located in the Great Lakes area), connecting cement plants to CO₂ pipeline networks is expected to be more challenging for facilities in remote locations. Pipelines can also face public resistance, and alternative means of transportation could be envisioned for the long-distance transportation of CO₂ such as rail and barges integrated with trucking for increased flexibility close to the capture and delivery locations.

CO₂ can also be used as a feedstock for building material processes, including curing concrete [47]. Utilization options might be financially attractive but present the risk to release CO₂ into the atmosphere later on and/or to create a product that is emitting CO₂ itself (e.g., oil from EOR operations). With the goal of decreasing use of fossil fuels, the most secure option for storing CO₂ long-term (> 1000 years) is injection in saline aquifers.

3.3.4. Cost

The cost of capturing CO₂ is inversely correlated with the concentration of CO₂ in the industrial exhaust stream. The exhaust streams from cement kilns and blast furnaces contain about 14–33 wt% CO₂ and 20–25 wt% CO₂, respectively [17,74,75]. As the cement kiln designs combine the process emissions and the emissions from fuel combustion, the exhaust stack might contain more pollutants that will have to be separated upstream of the CO₂ capture, increasing the cost of capture. The global CCS institute estimates the cost of CCS at cement and iron and steel plants in the U.S., including the cost of transportation and storage. First-of-a-kind costs should be roughly \$124/tCO₂ for cement kilns and \$77/tCO₂, for blast furnaces. This is expected to decrease for nth-of-a-kind infrastructure to roughly \$100/tCO₂ and \$65/tCO₂, respectively [101,102]. The competitiveness of CCS compared to heat provided by zero-carbon electricity or hydrogen based on zero-carbon electricity depends on the cost of electricity. According to a McKinsey report [103], the tipping point is \$50/MWh for the cost of zero-carbon electricity, over which CCS is more economically viable.

3.3.5. Opportunities in the U.S.

Fig. 8 shows the distribution of cement, lime, glass, and BF facilities in the U.S. along with CO₂ pipeline transportation and storage opportunities. Carbon capture retrofit on flue gas exhaust is a significant capital cost, so facilities that are expected to retire in the next decade are not expected to invest in that type of technology and thus were removed from the map. The best opportunities for CCUS in the short term are those facilities that are close to existing CO₂-EOR operations or close to CO₂ pipelines that are currently sending CO₂ to oil fields. This includes facilities located in Texas, New Mexico, Colorado, Oklahoma, Michigan, and Indiana.

Clusters of cement, lime, glass, and BF facilities can be found in many locations including close to Detroit (Michigan), Chicago (Illinois), St Louis (Missouri), Cincinnati (Ohio), Birmingham (Alabama), Los

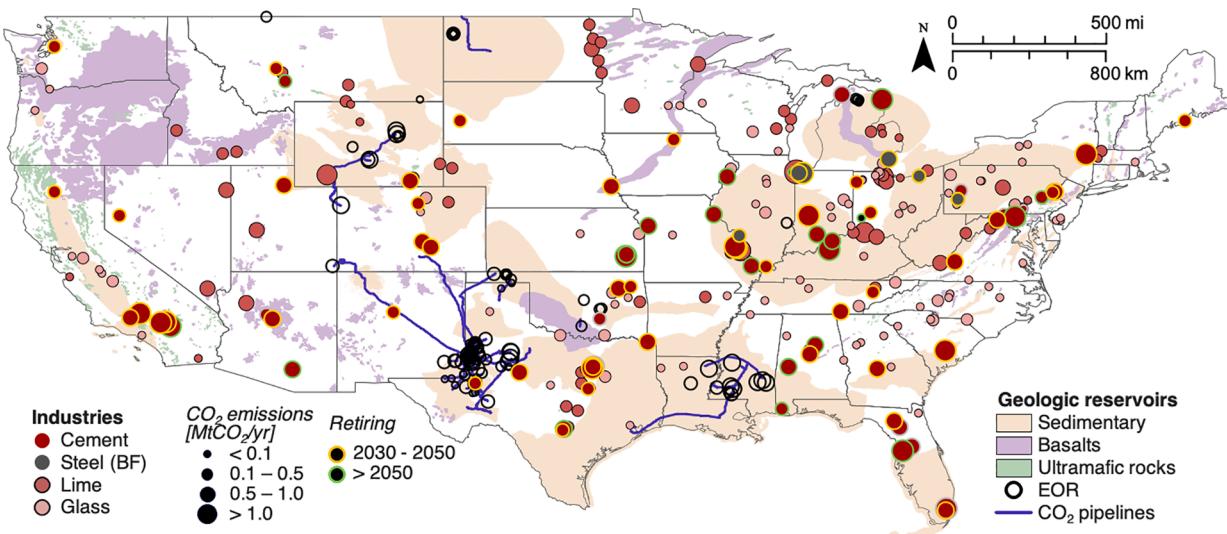


Fig. 8. Carbon capture in the cement, lime, glass, and steel industries, transportation, and storage opportunities in the U.S. for facilities retiring in more than 10 years [18,22,33–38,106–112]. Note that no retirement year estimate is given for lime and glass.

Angeles (California), Dallas (Texas), San Antonio (Texas) and Martinsburg (West Virginia). Each of these clusters are opportunities to find common places to sequester or utilize – depending upon the local context and the availability of geologic formations suitable for CO₂ sequestration – and to build pipelines that could connect nearby facilities to storage or utilization sites. For instance, the presence of a cluster of 4 cement plants and multiple lime and glass facilities in southern Indiana could justify the construction of a pipeline that would send the CO₂ to the EOR site located in Illinois. Clusters that are located over or close to sedimentary reservoirs but far from EOR operations would have to build dedicated CO₂ injection sites, like in southern Illinois, southern California, Pennsylvania, Alabama, or Texas. Blast furnaces are also mostly aggregated close to Chicago (Illinois), Detroit (Michigan), and Cleveland (Ohio) along with lime and glass facilities. If blast furnaces need to continue producing for supplying high quality steel, opportunities can be found nearby for CO₂ sequestration.

In addition, a study from the CO₂-EOR deployment working group [94] worked on a project regarding a CO₂ pipeline that would connect ethanol plants in the Midwest to the existing pipeline network in Texas and New Mexico. Three cement plants in Nebraska and Iowa are in the vicinity of this project and could be connected to the pipeline.

It should be noted that the implementation of CCS, pipeline transport, and storage is also dependent on the public perception of these projects. This has been shown in cases where public protests have led to CCS projects being canceled. An example of this was Shell's Barendrecht project in the Netherlands in 2010. Due to community activism and protest, regarding the fear that residents had about safety and housing prices, among other complications, the project was canceled [104,105]. Similar examples can be seen with the Vattenfall project slated in Beeskow, Brandenburg, Germany. Vetoos from Schleswig-Holstein and Lower Saxony concerning the CO₂ pipeline that would be used to transport CO₂ from local power plants put an end to onshore CCS for Germany [105]. Both of these cases resulted in the understanding that enhanced education and communications with surrounding communities and the public are needed when developing CCS projects.

3.4. Fuel switching opportunities

3.4.1. Replacement of coal with biomass

Fuel switching using biomass can be attractive for any facility, regardless of their next planned upgrade, as the utilization of some types of biomass do not require any infrastructure change. Drop-in biomass could be used right now to replace part of the coal and the conversion

factor could be increased in the future by using pre-processed biomass. Using biomass instead of fossil fuels in industrial kilns and furnaces does not necessarily reduce the emissions of CO₂ at the facility level; however, the source of CO₂ differs, which changes the lifecycle emissions of the feedstock and process. Fossil fuels source their carbon from the subsurface where it was securely stored for millions of years resulting in a net release of CO₂ in the atmosphere, whereas biomass sources its CO₂ from the atmosphere via photosynthesis. The full lifecycle emissions of biomass depend on the counterfactual use of the biomass, processing, and transportation to the facility [113–115]. For the purposes of this analysis, we assume biomass feedstocks would be carbon neutral. However, in practice there would be emissions from transportation and processing. Combusting biomass instead of coal thus reduces the emissions of fossil CO₂ in the atmosphere and can at best result in a carbon neutral cycle.

The cultivation of dedicated biomass feedstocks may require a significant amount of land, water, and energy and could disturb existing ecosystems and compete with other human needs such as food production and settlements. The use of residual biomass (e.g., wood residues, nut shells, and fruit pits) instead of whole trees can both protect intact ecosystems by reducing direct and indirect land use change and reduce reliance on artificial inputs. Land use change for dedicated energy crops might result in larger GHG emissions than the fossil CO₂ avoided by switching from coal to biomass in these industries. The type and source of biomass should thus be carefully considered to truly address climate change. The U.S. used about 330 million dry metric tonnes (Mdt) of biomass resources in 2017, including 130 Mdt of agricultural residue, for the generation of heat and power and the production of biofuels, biobased chemicals, and wood pellets [116]. The amount of unused agricultural residues in 2017 is estimated to be 95 Mdt, and is expected to rise gradually to 160–180 Mdt in 2040 [116]. For the purpose of this case study, we consider two types of biomass residue for fuel switching. First, we assess fruit hulls/pits and nut shells that are separated from edible crops at industrial processing facilities as secondary byproducts that have few existing use cases or markets despite their high energy density [117]. Second, we consider mill and forest management residues, including orchard prunings, unused mill residues, urban wood waste, and fire treatment thinnings [116]. In each of these cases, we reserve eligible biomass to feedstocks that would have been removed from the ecosystem or orchard regardless of the fuel-switching approaches described here and currently lack high volume and value use cases.

The heating value, carbon content, moisture content, and pore

structure distribution of a fuel influences its combustion. Substituting current fuels requires finding alternative fuels with similar characteristics. While sub-bituminous and bituminous coals are used in cement and lime industry, only bituminous coal is used in the steel industry [118] indicating the need for a higher quality coal. Given its characteristics, nut shells and fruit stones are particularly promising to replace solid carbon fuels (namely coal and petroleum coke) in some instances without any pre-processing, whereas woody biomass has a higher moisture content which might hinder the combustion (Table 4).

Some forms of biomass require more pretreatment than others before being usable as fuel for a given process. The pretreatment required is often pyrolysis, which reduces moisture content, improves grindability, and increases fixed carbon content [119]. The product of biomass pyrolyzation, biochar, is similar to coal in regard to its carbon, sulfur, volatiles, and ash contents. Additionally, pyrolyzation changes the biomass pore structure to be more comparable to that of coal [120–125]. The composition of coal compared to different types of biomass-derived fuel can be seen in Table 4, with additional properties listed in Table S1.

In the U.S., 11 cement plants are already co-firing biomass providing up to 1.6% of the heat generated [18,148]. Cement facilities currently use regionally available biomass: nutshells are used for heat generation in Arizona, California, Florida, and Texas, and corn is used in one facility in Iowa [148]. A facility in Oklahoma is also firing biogas in its cement kiln [148]. The geographic source of biomass is an important parameter and using local biomass would minimize the energy required for transport and the GHG emissions associated with it. Currently, high-quality biomass such as nut shells and fruit stones can be co-fired in cement and lime kilns with a maximum substitution of 20% on a thermal energy basis [149]. Because of the heterogeneity of biomass feedstocks, substitution beyond this rate may disrupt the stability and temperature of flame without changes to the kiln, potentially yielding inferior products [149].

Biomass has the potential to greatly reduce emissions intensity of steelmaking in the integrated mill production route, and this is currently being tested at the experimental scale [150]. It can be used in coke making at rates between 2% and 10%, resulting in an emissions reduction of up to 0.1 tCO₂eq/tonne steel. Biomass can also be used in the blast furnace as a substitute for nut coke (coke of smaller grain size) at a rate of 50% to 100%, resulting in emissions reductions of up to 0.2 tCO₂eq/tonne steel. It can be injected into the blast furnace in place of pulverized coal or natural gas injection to a rate of 25%, reducing emissions intensity by 0.5 tCO₂eq/tonne steel. Finally, it can also be used as a solid fuel in sintering, reducing emissions by 0.3 tCO₂eq/tonne steel. In all, emissions reduction opportunities of using biomass in the integrated steel production route total over 1.2 tCO₂eq/tonne steel, or nearly 60% of the emissions intensity of an integrated steel mill [150]. However, the production of coke does not always take place at integrated steel mills, so when determining the emission reduction potential from co-firing biomass, rather than including those from coke making and sintering, it was calculated solely for the BF and estimated to roughly 25% of its emissions.

Table 4
Composition of biochar derived from different types of biomass compared to the composition of coal.

Material	Fixed Carbon Content Before Pyrolysis (%)	Carbon Content After Pyrolysis (%) ¹	Moisture Content Prior to Additional Drying (%)	Higher Heating Value (MJ/kg)	References
Bituminous Coal	84 median (69–86)	N/A	8–18	24.4–32.6	[120,122,123, 126]
Subbituminous Coal	35.7–36.4	N/A	25.8–28.4	19.9–20.5	[120,122,123, 126]
Woody Biomass	12.85–18.–1	81.2–86.9	12.5–18.5	19.5–21.0	[39,127–129]
Nut Shells ²	16.3–27.6	77.3–87.4	3.3–10.5	17.4–20.6	[125,130–137]
Fruit Stones ³	19.2–23.8	84.0–89.5	6.2–35	18.8–21.4	[138–147]

¹ Temperature range for pyrolysis is 700–773 K, based on maximum energy density and yield tradeoff.

² Almond, hazelnut, walnut, pistachio, peanut, and pecan.

³ Apricot, plum, avocado, nectarine, peach, cherry, and olive.

Agriculture and forest residues are unevenly distributed across the U.S. While facilities in California, and in the Southeast U.S. might have multiple sources of biomass available, facilities in the Mountain West region might have greater difficulties sourcing local waste biomass in sufficient quantities to replace fossil fuels. While nut orchards process their crops mostly on site, fruits are more likely to be processed at beverage or canning facilities. This affects the strategy for purchasing these biomass byproducts. Moreover, there are several practical concerns for collecting and transporting biomass, including the volume of stones or shells removed at the commercial scale during processing and the current uses and commercial prices for these resources. In the next paragraph, we quantify the total technical potential of nutshell and fruit stones feedstocks, without considering collection constraints, alternative uses, transportation, or pre-processing. However, in practice these factors will determine the emissions abatement potential and economics of agricultural residues as kiln feedstocks.

Using the total annual tonnage of each type of biomass produced, adjusted to consider that the shell or stone only accounts for a portion of the mass, it was determined that the U.S. produces 1,488,000 tonnes of nut shells and 241,700 tonnes of fruit stones [120,124,125,130–135, 137–147,151–179]. The heating values of the nut shells range from 17.4 to 20.6 MJ/kg, and the heating values of the fruit pits are between 18.8 and 21.4 MJ/kg [120,124,125,130–135,137–147,151–179]. These heating values and the quantities of biomass were used to calculate the available energy from these biomass sources, finding that nut shells and fruit stones could provide 2.93×10^{10} MJ/yr and 4.70×10^9 MJ/yr, respectively (feedstock volumes and shell/stone percentage of biomass total mass, available in Table S1 in the Supplementary Information). These figures can be compared with the total energy provided by coal and coke in the cement and steel industries, that equal approximately to 2.16×10^{11} MJ/yr and to 1.91×10^{11} MJ/yr, respectively [148,180]. The available energy from the waste nut shells account for 7.2% of the energy used in these industrial processes and fruit stones 1.2%. The share of energy from coal and coke in the lime industry and the emissions intensities were assumed to be the same as in the cement industry given the similar processes. This indicates a consumption of roughly 5.56×10^{10} MJ/yr in the lime industry. The replacement of coal by nut shells and fruit stones could reduce fossil CO₂ emissions by roughly 2.7 MtCO₂/yr (1.8% of emissions from cement, lime, and primary steel-making in the U.S.) in the U.S through direct substitution on a thermal basis [18,32]. This does not account for the other aspects of using biomass, which could reduce this potential. For example, charcoal-based steel production has a lower efficiency than coal-based steel production (about 60%) [103].

Fig. 9a shows the distribution of nut and fruit production along with the distribution of cement, lime, glass, and steel plants [18,106,107, 155]. The amount of coal that could be replaced by drop-in biomass in cement kilns, lime kilns, and BF is calculated at the state level on an energy basis. Details are given in the SI regarding the emissions from fuel combustion, the share of coal in the fuel mix, and the share of coal that can be replaced by drop-in biomass. This fuel substitution has the

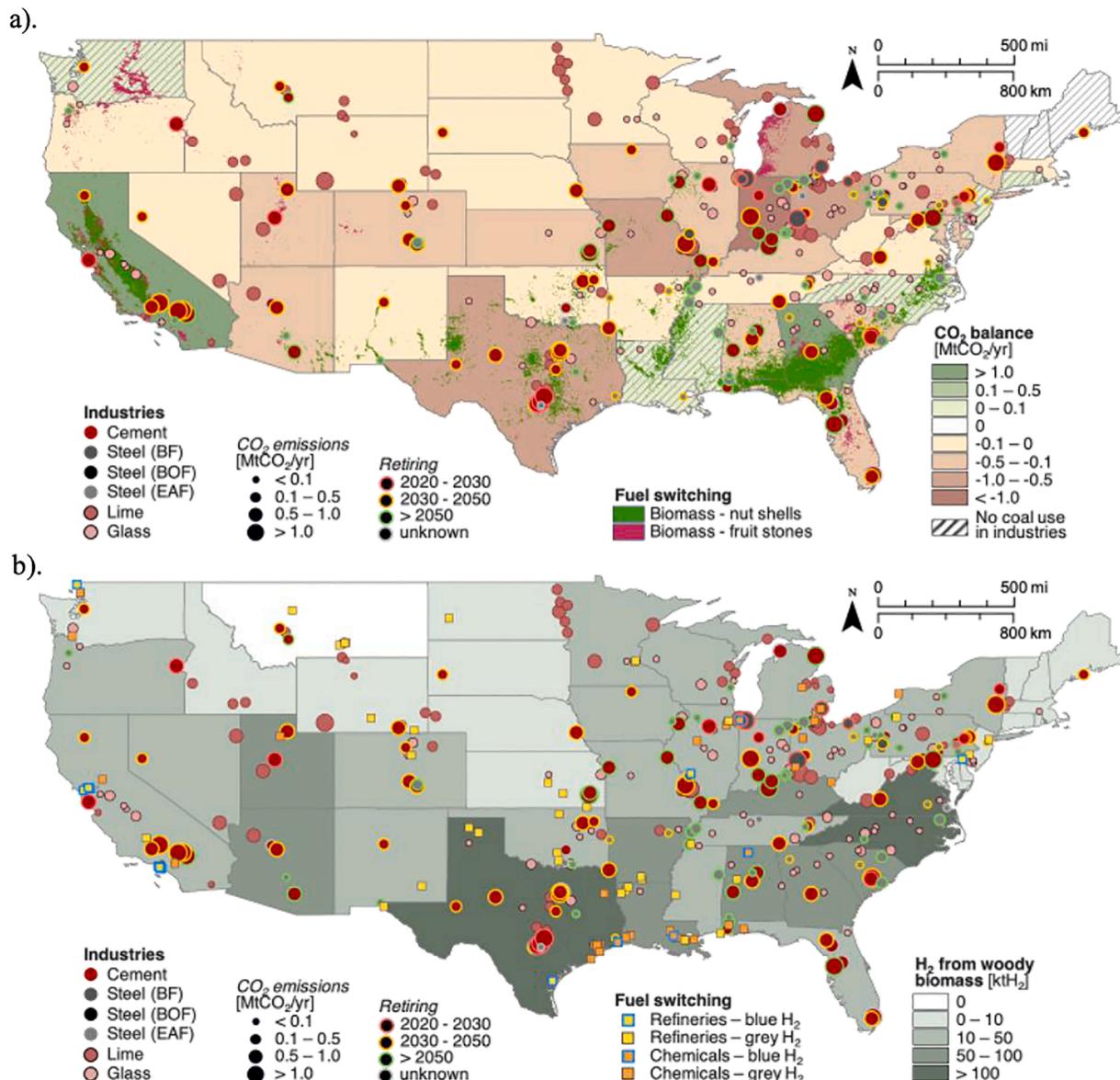


Fig. 9. Opportunities in the U.S. to fuel switch using biomass and hydrogen in the cement, lime, glass, and steel industries. The top map shows the CO₂ emissions that could be offset by replacing 20% of coal with drop-in biomass, indicated by the CO₂ balance. The green colors indicate an excess of drop-in biomass compared to the needs in BFs, cement, and lime kilns while the orange-red colors indicate a deficit in nut shells and fruit pits availability, showing the need for other types of biomass sources or other types of decarbonization options. The bottom map shows the hydrogen that can be produced from waste woody biomass sources [18,106,107,116, 155,192].

potential to result in an overall fossil CO₂ emissions reduction of 4.3 MtCO₂/yr (6.4% of annual emissions), 1.1 MtCO₂/yr (3.6% of annual emissions), and 3.1 MtCO₂/yr (5.8% of annual emissions) at cement kilns, lime kilns, and blast furnaces, respectively [18,32]. However, the availability of the drop-in biomass feedstocks identified above would not be able to cover the needs of that scenario and could only offset up to 2.7 MtCO₂/yr.

The energy provided by coal and coke was compared to the energy potentially available from nut shells and fruit pits for each state and the associated CO₂ emissions reduction. The calculations assume that the biomass is harvested and used in the same state. This analysis highlights the states that would have to look for other types of locally available biomass, which might require additional pre-processing, or other types of alternative fuels. Nut shells and fruit stones are unevenly distributed with about 830,000 dry tonne (dt) biomass in California, 350,000 dt in Georgia, 150,000 dt in Florida, and 90,000 dt in Alabama. This distribution positions California, Georgia, and Florida as regions that could

utilize drop-in biomass at cement, lime, and steel plants. However, most of the blast furnaces, which would also benefit from biomass fuel switching, are located in the Great Lakes region, requiring the biomass to be transported for utilization, and posing additional difficulty. Enough nut shells are available in California and Georgia to replace 20% of the coal in the cement plants as recommended by Murray and Price [149].

3.4.2. Replacement of fossil fuels with hydrogen

In regions where waste biomass is more difficult to acquire locally, hydrogen may be a viable replacement for carbon-intensive fuels in kilns and furnaces. Today the production of hydrogen is still tightly related to fossil fuels, as 95% of the hydrogen is produced via steam methane reforming (SMR) in the U.S. [181]. Worldwide, 48% of the hydrogen comes from SMR, 30% from petroleum reforming, 18% from coal gasification, and 4% from water electrolysis [182]. Future clean sources of hydrogen include SMR with sufficient (90%+) carbon capture, metallic

membranes and water electrolysis [183]. Hydrogen from these sources are often referred to using different colors; gray for hydrogen that comes from natural gas, such as through SMR ($\sim 355 \text{ gCO}_2\text{eq/kWh}$), blue for hydrogen from natural gas with sufficient carbon capture ($\sim 40 \text{ gCO}_2\text{eq/kWh}$), and green for hydrogen produced through water electrolysis using renewable electricity ($\sim 14 \text{ gCO}_2\text{e/kWh}$) [184]. The use of hydrogen fuel switching has gained the attention of the scientific community but has yet to be proven at the industrial scale for cement, lime, and glass. It has, however, gained traction in the iron and steel industry for DRI. DRI is usually achieved in a carbon monoxide-rich environment, resulting in pure iron and CO_2 . Instead, a hydrogen environment can be used with only iron and water as resulting products [185].

There are many initiatives and pilot programs in the steel industry aimed at developing hydrogen iron ore reduction and melting technology. HYBRIT is a pilot project which began in 2016 in Sweden – headed by steelmaker SSAB, iron ore mining company LKAB, and gas provider Vattenfall—that aims to demonstrate nearly zero-carbon, fossil-free steelmaking using hydrogen. Construction on the pilot plant began in 2018, and the companies indicate that initial costs of steel production to be 20% to 30% higher than fossil fuel-based steel production, but that this cost gap could shrink as the technology is improved [186].

Assuming an energy density of 121 MJ/kg hydrogen gas, approximately 80 kg of hydrogen is needed to produce 1 tonne of steel [186]. Therefore, a 5 million tonne/year steel plant would require 400,000 tonnes of hydrogen per year, or approximately 45 tonnes of hydrogen per hour. Current global production of hydrogen is around 70 million tonnes [187], which, if fully utilized for steelmaking, could produce 175 million tonnes of steel, or approximately one tenth of current global steel production. As renewable energy production is added to electricity grids, dedicated and excess renewable energy could be used for hydrogen production by means of electrolysis.

Additional considerations and research are needed to validate the successful use of hydrogen in cement, lime, and glass kilns. Hydrogen burns at much higher temperatures and has a higher flame speed than the fossil fuels used in these applications today, so material requirements and burner configurations for the kilns might be altered upon adoption of this emissions-reduction pathway [188]. By changing the burner configuration to one that promotes a diffusion flame upon the adoption of a hydrogen-fired system, additional NO_x emissions from the higher burn temperatures can be reduced [188]. Coinciding the adoption of this technology with an update and relining could better justify the cost of the upgrade to a hydrogen-fired system.

Hydrogen flames burn differently than fossil fuel powered flames, specifically with less soot production. The reduction of soot promotes less particulates in the combustion environment and the exhaust gas, but also reduces the radiative heat transfer available within the kiln [189]. The reduced radiative heat transfer might be made up by the higher temperatures at which hydrogen burns. These differences would require significant changes to existing processes to accommodate for this change in fuel [12].

When hydrogen is used to produce cement, lime, glass, or in the blast furnace, there will still be CO_2 process emissions in the exhaust stream. However, the reduction of soot in the exhaust gas is beneficial because it will reduce the risk of poisoning any carbon capture module that would be implemented. The major constituents of this exhaust gas would be CO_2 and water vapor. The effects of the newly present moisture would need to be evaluated such that they do not interfere with the quality of the clinker, lime, glass, or iron products. Before hydrogen can be used for full decarbonization of cement, lime, and glass kilns or blast furnaces, it could be used as an additive to existing fuel for significant emissions reductions [13]. The CEMEX plant in Spain recently finished a campaign in 2019 in which it was fired with a blend of natural gas and hydrogen. CEMEX plans to keep iterating on this process, with the goal of reaching 5% hydrogen injection by 2030 [190]. The company plans to test a plant that will run on 100% hydrogen starting in 2021 [191].

Current sources of hydrogen are hydrogen plants via SMR, petroleum

refineries, and biomass. Hydrogen is produced through various chemical reactions including gasification of fossil fuels or biomass and electrolysis of water. In the U.S., 12 refineries out of 55 and 5 hydrogen plants out of 57 are suppliers of CO_2 according to the EPA [18]. This means that 17 locations producing hydrogen already have CO_2 capture systems and can supply blue hydrogen and 75 plants can supply gray hydrogen. Ideal production places would be the Great Plains for wind energy and California, Texas, and the Southeast for solar energy, even if multiple other opportunities exist. As described above, the use of waste woody biomass for sustainable hydrogen production has U.S. wide opportunities.

Due to its lower carbon content and higher moisture content (see Table 4), woody biomass is a good candidate for bio-syngas generation. About 13 kg of biomass are needed for generating 1 kg of hydrogen [192]. Oak Ridge National Lab (ORNL) made the inventory of biomass in the United States and in 2016 forest residues, forest thinnings, primary mill residues, and secondary mill residues totaled approximately 20 million dry tonnes of biomass [116]. This could result in the production of up to 1.5 Mt H_2 , that could be used in multiple industries for producing heat. Fig. 9b shows cement, lime, glass, and steel facilities, the readily available waste biomass and hydrogen, and the estimated amount of hydrogen that could be produced from the waste biomass in each state.

3.5. Technological changes

3.5.1. Fluidized bed reactors

When kilns need to be replaced, they are often replaced by the same technology. However, with the substantial investment of time and money needed to fully replace a retired kiln, upgrading to a new technology that promotes emissions-reduction should also be considered a viable option. One of those technologies is a fluidized bed reactor. As shown in Fig. 10, feedstock enters the fluidized bed at the top of the reactor while hot gasses are fed in towards the bottom. Additional air is fed in at the bottom of the reactor at the fluidization velocity causing the particles and hot gasses to behave like a liquid [193]. The increased combustion efficiency and heat transfer in a fluidized bed reactor allows for shorter processing time and more effective use of fuel. The only limitation to the hot gas composition is any that would compromise the end product. This allows for the hot gasses to be optimized using waste gas or a specific blend to reduce the energy needed for carbon capture. Fluidized bed reactors require more electrical input than rotary or shaft kilns due to the blowers needed to provide the fluidizing air. Electricity sourced from low-carbon resources can meet this demand without compromising the emissions reductions gained from this technological innovation.

Fluidized bed reactors can be used in the cement, and lime industries to heat the feedstock materials. The limestone and clay feedstocks must be preprocessed to ensure they are finely ground before entering the fluidized bed kiln. The clinker and lime products that are recovered from the fluidized bed kilns are typically smaller (1–3 mm in diameter) than those that are formed in a rotary or shaft kiln [194]. Fluidized bed reactors can also be applied in iron reduction. The iron ore is usually pelletized or finely ground before use in a fluidized bed, but the preparation can vary depending on the technology.

The Japan Coal Energy Center (JCOAL) has developed the Fluidized-bed Advanced Cement Kiln System (FAKS) and has demonstrated its ability to produce 1000 tonnes of clinker per day. The configuration tested at this capacity from 2005 to 2008 was fueled with coal and resulted in a decrease of 10,000 t CO_2 /yr (8.5% of annual CO_2 emissions compared to the baseline case) [197]. JCOAL reports that FAKS is expected to be adopted on the commercial scale for cement production. Improvements can still be made by switching the coal fuel to natural gas and by partnering FAKS with CCS. Additionally, in the US, Specialty Minerals uses a fluidized-bed kiln to produce lime in Adams, MA [22]. Given the similarities between the cement and lime industries, the technological development and learning from one industry may be

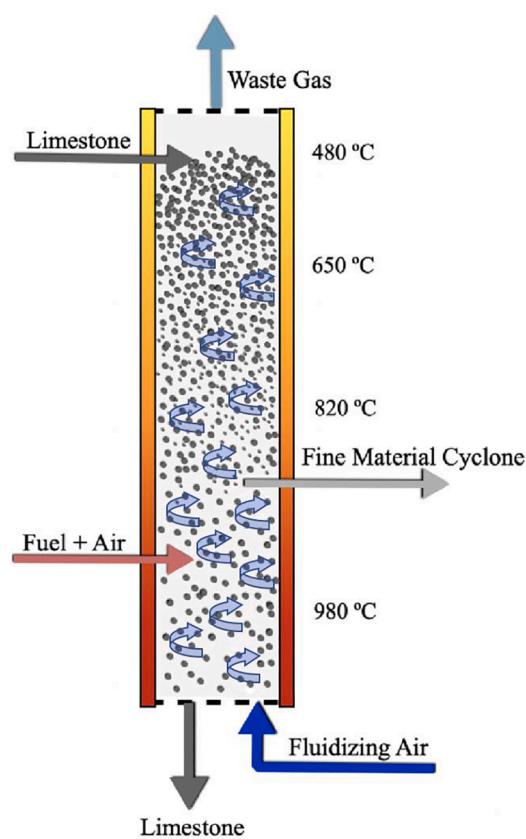


Fig. 10. Representative schematic diagram of a fluidized bed furnace using limestone. In this schematic, the kiln temperature is maintained via combustion of a given fuel with air, resulting in a waste gas that exits from the top of the kiln. The fluidizing gas is air which is used to ensure the proper residence time for the limestone inside the kiln. The calcined limestone will exit through the bottom of the reactor as a product, while the limestone particles too fine to use are pulled from the calciner to a fine materials cyclone which separates and treats these byproducts [195,196].

transferable to the other [198].

Fluidized bed technology in the steel industry was pioneered in the 1990s with the invention of the FINMET process. This provided a continuous production method of hot-briquetted iron from iron ore fines, which offered a cheaper input relative to pellets and iron ore lumps. There are no FINMET plants currently in production due to the limitations of the high pressures that were required, but the FINEX process, developed by Primetals Technologies and Posco, combines the original FINMET process with COREX melter-gasifier technology to result in a continuous production process that uses iron ore fines as input and produces hot metal equal in quality to blast furnace technology [199,200]. A FINEX plant in Korea with a production capacity of two million tonnes per year commenced operation in 2014 [201]. Additionally, Metso Outotec has developed a fluidized bed technology, Circored, that is used to produce DRI and hot-briquetted iron for direct use in an EAF [202]. This system is powered by natural gas, but the reduction is primarily hydrogen-based. Given the Circored process coupled with an EAF CO₂ emissions can be reduced as much as 90% in comparison to a conventional BF-BOF route [202]. In 2010, Metso Outotec also expanded their facility to incorporate a CO₂ removal pilot plant that can be paired with the iron reduction fluidized bed [203].

3.5.2. Oxyfuel kilns and furnaces

In oxygen-fired (also referred to as oxyfuel or oxycombustion) kilns the main difference is that the air in existing kilns and furnaces is replaced by an enhanced oxygen environment, often requiring an air

separation unit (ASU) to be located onsite. Replacing air with oxygen results in higher flame temperatures and higher temperatures of the exhaust gas, which could reduce overall energy requirements and process time. By removing the nitrogen, the thermal efficiency is increased to nearly 80%, while for air it is between 40–60% [35–37]. When fossil fuels are used in an enhanced oxygen environment, the exhaust gasses mainly consist of CO₂ and H₂O, as opposed to exhaust gas from an air environment, which also includes N₂ and NO_x. The CO₂ and H₂O mixture requires less processing and energy to separate the CO₂ and can be achieved through a condensing unit, which is often much less expensive than conventional carbon capture equipment. For oxy-combustion with fossil fuels to result in avoided CO₂ emissions, it must be coupled with CCS.

An oxyfuel glass furnace was first demonstrated by the Carr-Lowrey Company in 1990 using an 73-tonne-per-day (TPD) furnace. Soon after this demonstration, the Gallo Glass Company, the largest glass producer in North America, permanently converted one of their 318 TPD furnaces to operate in oxyfuel conditions [38]. Since then, oxyfuel furnaces have been implemented at industrial scale throughout the glass industry. In oxyfuel conditions, glass requires less energy for production and the NO_x emissions are reduced. The main considerations when glass producers switched to oxyfuel technology were the refractory materials and the geometry of the hearth kilns [39]. To further the adoption of this technology, the U.S. DOE hosted two workshops focused on the changes that would need to be made to successfully adopt this technology [38].

Oxyfuel kilns can also be applied to the cement and lime industries, as well as the blast furnace in ironmaking and electric arc furnace in steelmaking. The basic oxygen furnace already employs this technology and could be connected to a downstream CCS unit to result in avoided CO₂ emissions. To deploy oxyfuel in the cement industry, there needs to be considerations surrounding the precalciner cyclones that utilize the exhaust gas to preheat the feedstock. The elevated concentration of CO₂ in the exhaust gas could lead to less feedstock decomposition in the precalciner cyclones, however this can be remedied through alterations in process infrastructure [35]. In 2009, LafargeHolcim, FLSmidth, and Air Liquide collaborated to design and test a pilot scale oxyfuel calciner. The FLSmidth pilot plant calciner was modified to be oxy-fired and 7 test campaigns were run with a feed rate of 2 t/hr through 2011 and 2012 [40]. These partners concluded that there were no safety concerns with the oxyfuel process, there was no impact on the quality of cement that was produced, and existing preheaters and calciners can be retrofitted for oxycombustion. Due to the similarities between the cement and lime processes, the technological innovations in the cement industry are expected to be transferable to the lime industry.

Pure oxyfuel technology has not yet been demonstrated in iron and steel blast furnaces, but experiments have been conducted to evaluate the feasibility and advantages of adding oxygen enrichment. A full oxygen blast furnace (FOBF) would still allow for coke to be used as a fuel such that the carbon monoxide environment will still reduce the iron ore. The main hurdle faced in deploying a FOBF is the inconsistent temperature profile that would result in a “hot bottom and cold top problem” [37]. To mitigate this problem, the blast furnace top gas can be recirculated back into the furnace with fuel input. This recirculation also increases energy efficiency. In addition to conventional oxyfuel technologies that can be adopted for use in the blast furnace, flameless oxyfuel technology can provide more stable environments for the use of blast furnace top gas farther down the iron and steel production line [37].

3.5.3. Electric kilns and furnaces

The use of electric kilns and furnaces can help reduce process and stationary combustion emissions in industrial processes. This is seen in the use of an EAF for steel production, where process GHG emissions are only due to combustion of fuels for process heating, indirect emissions due to electricity usage, and the degradation of the graphite electrodes [32]. EAFs are most commonly used in secondary steel production,

through which new steel products are made from recycled steel scrap, as opposed to primary steel production where steel products are made from virgin iron ore. EAFs, therefore, rely on the availability of steel scrap, and the quality of steel produced in the EAF is largely determined by the quality of steel scrap used in production. Steel scrap quality is characterized by the number and amount of impurities contained in the scrap, such as copper, sulfur, tin, phosphorus, and zinc. Higher quality scrap can be used to produce automotive steel, while lower quality scrap can be used to make rebar and other structural steel products. Given the limited supply of high-quality scrap, EAFs are predominantly used in the production of structural steel products, where corrosion resistance and other special physical qualities of the product are not required [204].

EAFs can also use direct-reduced iron, pig iron, and hot-briquetted iron as feedstocks, and these can be used to increase the quality of the feedstock by diluting the impurities in the scrap. The DRI-EAF production method can, in theory, be used to produce any quality of steel, including exposed automotive sheet, as the DRI can be tailored to contain any specified alloying elements and purity of iron [204,205] (see Fig. 11). Globally, DRI production exceeded 100 million tons in 2019 [206] so, the DRI-EAF production route is a proven technology that can be scaled up to produce high-quality steel products in the near future.

In steel EAFs, the electric arc can be formed between the electrode and the steel scrap or DRI since iron and most steels are electrical conductors. However, for a material such as limestone, an indirect arc furnace would need to be considered. An indirect arc furnace conducts an electric arc between electrodes, and the process material is heated through the radiant heat transfer from the arc [207]. This allows the indirect electric arc configuration to be used in the production of non-conducting materials.

The EAF is only one method of electrical heating. Other methods include direct and indirect resistance heating. Resistance heating can be used in the cement, lime, and glass industries. Resistance heating in a rotary kiln is achieved by heating the rotating steel drum on the outside with electric heaters, while the material is fed through the inside, as shown in Fig. 12. The inside of the drum is lined with refractory material

to assist in keeping the cylindrical shape under the high temperature conditions. IBUtec and FEECO are both prominent suppliers of indirectly heated rotary kilns, which tend to have lower material throughput than natural gas or coal fired rotary kilns [208,209]. As quoted by FEECO, indirect-fired rotary kilns have a reduced throughput of 91 kg/hr (200 lb/hr) to 18 tonnes/hr (20 tons/hr) [208], while IBUtec quotes 0.1 kg/hr (0.2 lb/hr) to 1000 kg/hr (2,200 lb/hr) [209]. The material throughput reduction is due to the inefficiencies of conductive heat transfer from the electric heaters to the steel drum, to the refractory, to the material, or to the kiln atmosphere. Additionally, the use of electric heaters also eliminates the radiative heat transfer that would be present given a flame.

Resistance heating is also used in the glass industry and is often referred to as electric boosting. The resistance heaters are submerged into the glass feedstock, and as they are heated, the glass melts around them and then flows out of the hearth furnace where it is cooled. In some existing configurations, the resistance heaters are positioned at the bottom of the hearth kiln extending upwards into the glass feedstock [210]. This allows for the furnace to be co-fired with natural gas, resulting in electric boosting or for all electric melting. Major considerations when switching to an all-electric melting system in a glass furnace are the positioning and the resulting convection currents in the molten glass [210,211]. The Fives Group and Electroglass have both developed all electric glass kilns for most types of glass production. These kilns have the ability to produce from 20 kg/day (44 lb/day) to over 250 tonnes/day (2755 tons/day), ranging from an energy intensity of 720 - 850 kWh/tonne of glass produced [212]. These configurations have resulted in increased energy efficiency and reduced CO₂ emissions.

Although the use of electric kilns and furnaces does not reduce the process CO₂ emissions produced by calcination, these processes could be paired with CCS to become carbon neutral. It must also be noted that to fully decarbonize the energy needed for these industrial processes via electricity, the electric grid or source of electricity must be decarbonized. Additionally, co-locating electric kilns with low- or no-carbon electricity can reduce or eliminate the CO₂ emissions from the electricity generation needed for cement and lime calcination or glass melting. Fig. 13 shows the areas in the U.S. where cement, lime, glass,

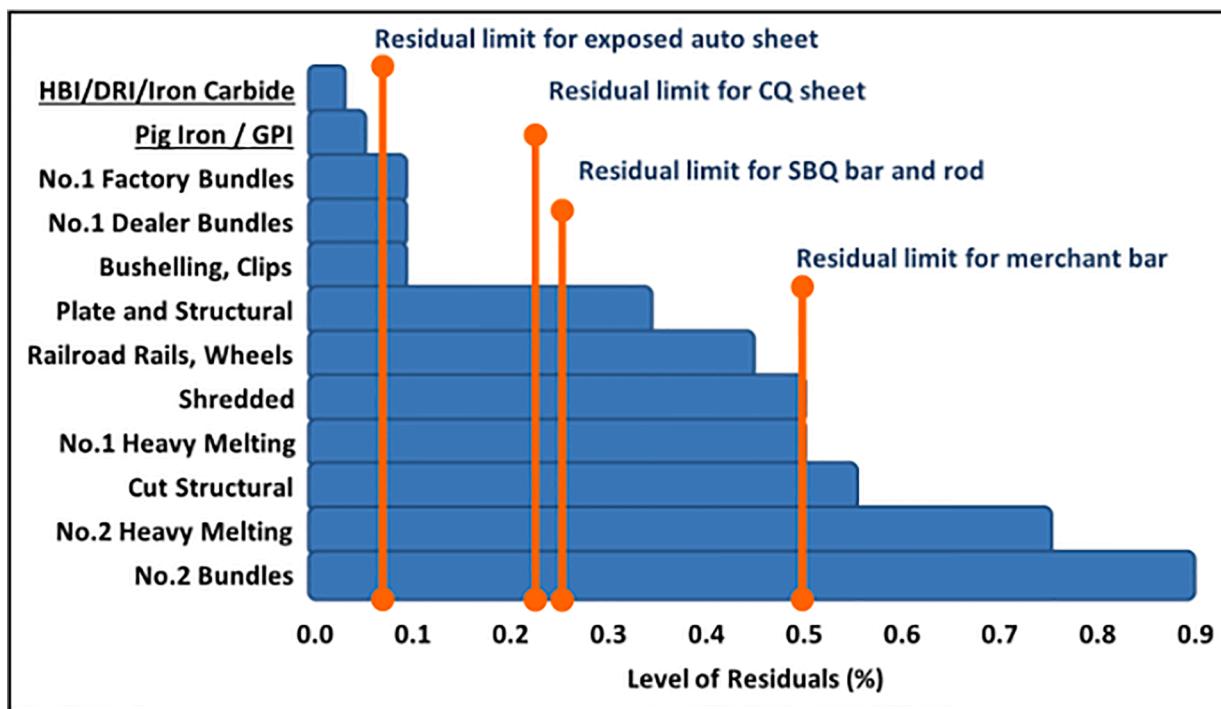


Fig. 11. Level of residual impurities allowed for various steel applications. Sourced from IIMA, *Use of Direct Reduced Iron (DRI) in the Electric Arc Furnace (EAF) for Steelmaking* [204].

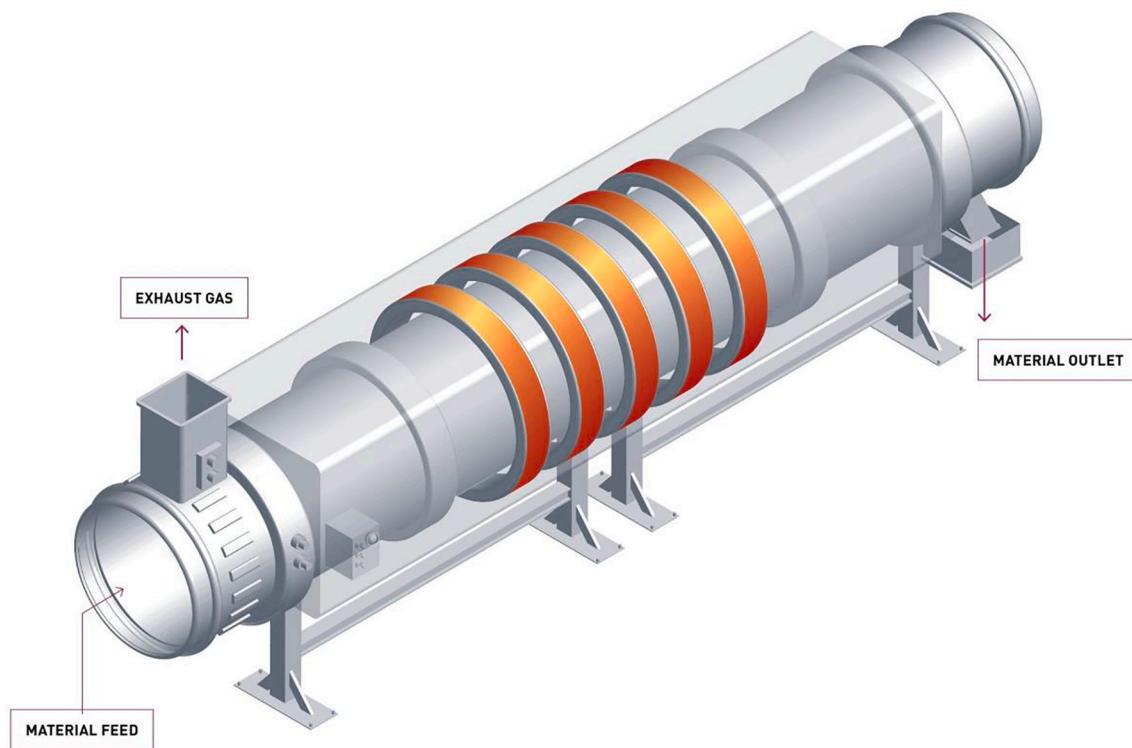


Fig. 12. Indirect resistance heating applied to a rotary kiln in which the resistance heaters are located around the rotating shell of the kiln. Image courtesy of IBUtech Advanced Materials, Weimar, Germany [209].

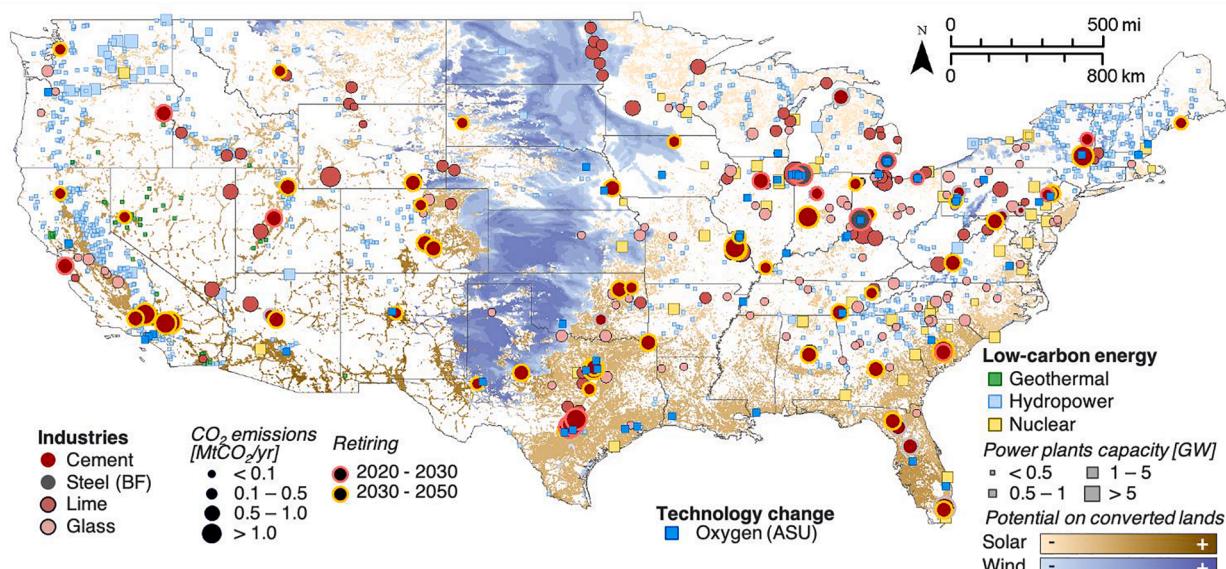


Fig. 13. U.S. map showing cement, lime, glass, and iron and steel facilities with retiring kilns in the next decades overlaid with the locations of no- or low-carbon energy and established air separation units (ASUs). The co-location of the retiring facilities and these resources can be evaluated to determine which of the innovative pathways may be most accessible and least costly to adopt upon kiln or furnace retirement [18,33–38,106,107,213–216]. The list of ASU locations might not be complete due to proprietary data.

and blast furnaces can be co-located with low- and no-carbon electricity sources so these facilities can take advantage of the natural resources that are available.

3.6. Other CO₂ reducing innovations

In addition to the emissions-reducing technologies covered

previously, there are other emerging innovations with the same aim. The Low Emissions Intensity Lime and Cement (LEILAC) project, located at the HeidelbergCement plant at Lixhe in Belgium, uses technology that decouples the exhaust gas of calcination from the exhaust gas of fuel combustion at cement plants in order to increase the concentration of CO₂ in the process exhaust and lower the cost of capture [103]. This pathway was developed by Calix Global and has proven productive

when calcining magnesite at their commercial-scale plant in Bacchus Marsh, Australia [217]. Their design allows their kilns to be heated in various ways, making it easier to couple to the most cost or emissions efficient energy source.

The Hoffmann GreenCement Technologies startup has developed a way of making cement that does not require any mining of raw materials or heat input. The process is based on the use of industrial by-products such as blast furnace slag, clay slurry and gypsum that is reacted with chemical activators. This reduces the CO₂ emissions by 5 to 188 kgCO₂ per tonne of cement [218]. The availability of industrial by-products might however limit the production of low-carbon cement with this process, as the steel industry is also looking for reducing its carbon footprint. This might result in the retirement of BF, and the decrease of blast furnace slag supply.

Origen Power is a company that is developing a fuel cell powered calciner. To achieve this, methane is fed to a fuel cell, which then produces electricity that is used to calcine limestone, producing lime and a pure stream of CO₂, which can be compressed for storage [219,220].

Another option that has been introduced to electrify cement production is electrochemical calcination. Electrochemical calcination involves splitting water and limestone (CaCO₃) into their component parts by applying renewable electricity. The hydroxide (OH⁻) and CaO combine to form Ca(OH)₂ (slaked lime) in solid form, while releasing H₂, O₂, and CO₂ in pure form. The slaked lime produced from this reaction can be used in the production of Portland cement, as well as other applications [221].

Electrolysis and electrowinning can be used to reduce iron ore to make steel. A U.S. research firm, Boston Metal, is leading the research and development of molten oxide electrolysis steelmaking technology. The electrolytic process uses electrodes and a liquid oxide electrolyte at a temperature of 2000 °C to separate iron from oxygen and other elements found in iron ore [222]. Additionally, the Siderwin steel project is a consortium of 12 companies whose aim is to build a pilot plant by 2022 demonstrating large-scale steel production by an electrowinning process [223]. The electrowinning process differs from the electrolytic process in the type of electrode and solution used. By eliminating the need for a blast furnace and coke, the Siderwin process is expected to reduce the energy use and carbon intensity of integrated steelmaking by 30% [224].

3.7. Combination of innovation pathways

CCS can be combined with many of the proposed innovative pathways towards decarbonization to result in carbon-neutral or carbon-negative operations.

3.7.1. Biomass and CCS

One of the main paths to reducing anthropogenic CO₂ emissions is to reduce the use of fossil fuels. As discussed above, biomass could replace coal in the cement and steel industry. Burning biomass still emits CO₂, but its source is different. While the source of CO₂ emitted by fossil fuels is the subsurface, biomass is sourcing its CO₂ from the atmosphere. Using sustainable biomass in cement kilns and blast furnaces with CCS is a means of capturing CO₂ from the atmosphere and could thus be considered as a type of bioenergy with carbon capture and storage (BECCS), which is traditionally propositioned to be used for the electricity production. Due to the emission of CO₂ as a byproduct of the cement and steel manufacturing, using sustainable biomass instead of fossil fuels whenever possible is the only path that could result in a carbon neutral process or in negative emissions. To verify the neutrality or negativity of a process that employs both biomass fuel switching and CCS, a lifecycle analysis must be conducted such that the less-carbon intensive process can be adequately compared to the baseline case using coal. Tanzer et al. (2020) has modeled multiple pathways to achieve emissions reductions in the steelmaking industry through the use of both and has concluded that with high rates of biomass fuel

switching and use of CCS, CO₂ emissions from steel production can be nearly offset [115].

3.7.2. Oxyfuel and CCS

One of the many benefits of an oxyfuel kiln is that the exhaust gasses consist of only H₂O and CO₂. To avoid the CO₂ emissions entering the atmosphere, this technology can be paired with CCS. This can be achieved by simply condensing the H₂O out of the exhaust stream, leaving high-purity CO₂. The CO₂ can then be pressurized for transportation and storage. The advantage of this combination of innovative pathways is that it leads to a separation process that is significantly less energy intensive and less expensive than CCS given an air combustion environment. Oxyfuel combustion can also be paired with a fluidized bed kiln configuration by replacing the fluidizing air with pure oxygen. This combination of innovative pathways would lead to the same separation process using a condenser, but also enhance the heat transfer to the feedstock material. By capturing the CO₂ from the oxyfuel kiln, the resulting operation can be close to carbon neutral.

3.7.3. Electricity and CCS

The use of electricity in industrial furnaces ultimately reduces the CO₂ emissions that would come from the combustion of fossil fuels, but it does not mitigate the process emissions associated with the heated feedstock. By combining electric furnaces with CCS, the CO₂ process emissions can be separated from the other exhaust gasses and pressurized for transportation and storage. Given a renewable energy source, the capturing of process emissions from an electric furnace results in a carbon-neutral operation.

4. Case studies

4.1. California and biomass fuel switching

4.1.1. Context

In California, GHG emissions totaled to 418.2 MtCO₂eq in 2018, 21.1% from the industrial sector [226]. In this state, 8 cement, 2 lime, and 6 glass facilities respectively result in 8.1 MtCO₂eq, 0.16 MtCO₂eq, and 0.5 MtCO₂eq of emissions [18]. The state also presents various options for CO₂ sequestration—deep saline aquifers in the Central Valley, basalts in the North East, and ultramafic rocks in the North West – and for fuel switching – large resources of biomass waste in the Central Valley, and hydrogen (SMRs) mostly close to Los Angeles and San Francisco (Fig. 14).

4.1.2. Carbon capture

Given this beneficial geology in California, carbon capture and storage should be considered for cement facilities who are not approaching retirement or kiln technology upgrading within the next 10–20 years. If each of the facilities that are projected to retire after 2030 were to adopt carbon capture and storage with a capture rate of 90% from cement kilns, 6.3 MtCO₂eq (77% of annual in-state cement emissions, 7.1% of annual in-state industrial emissions) anthropogenic emissions could be kept out of the atmosphere and stored permanently in geologic reservoirs, negating CO₂ emissions incurred from compression, transportation, and injection in the subsurface [65].

4.1.3. Fuel switching

Another option for all cement and lime facilities is fuel switching with biomass wastes. In the Central Valley of California, there are an estimated 731,000 tonnes of nut shells and 99,000 tonnes of fruit pits [124,125,130–135,137–147,151–179]. Roughly 40% of this resource could be used to replace 20% [48] of the 5.2 million tonnes of coal currently used in these industrial processes. This replacement could reduce anthropogenic emissions by 0.5 MtCO₂eq (6.2% of state emissions from cement and lime, 0.6% of annual in-state industrial emissions) compared to the current use of coal.

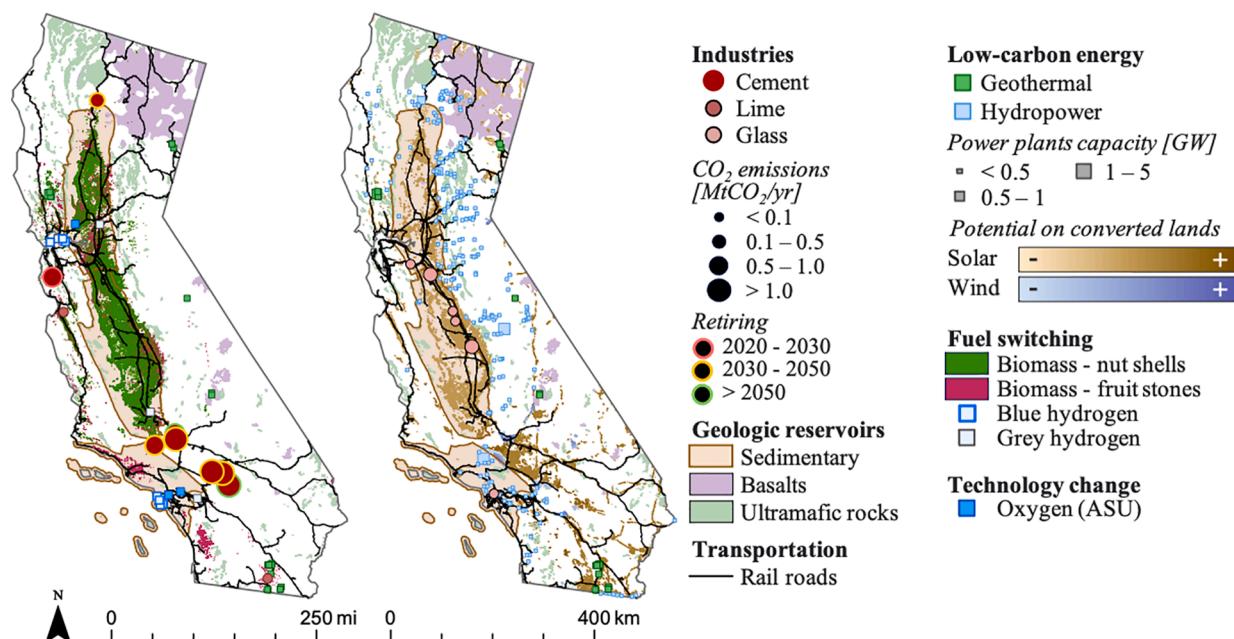


Fig. 14. Maps for California highlighting alternative pathways to coal use at cement and lime facilities by fuel switching using biomass or hydrogen or by technological change to oxyfired kilns using commercial-scale air separation units (ASUs) (left map) and the electrification of glass facilities using renewable sources of electricity (right map). These pathways are overlaid with CO₂ storage opportunities, and with the railroad networks to illustrate transportation potential [18,106, 108–111,155,213–216,225]. The list of ASU locations is not complete due to proprietary data.

4.1.4. Technology upgrade

For facilities that have retiring kilns or are in the market for upgrading their technology, hydrogen-fired, oxyfuel, and electric kilns should be considered. In central and southern west California, there are opportunities to leverage co-location near blue hydrogen (hydrogen production with CCS) sources for kiln or furnace fuel production in the Bay Area and close to Los Angeles. Additionally, in similar locations, large air separation units are stationed and could provide opportunities for partnerships given an oxygen-demanding oxyfuel kiln or furnace. Electric kilns should be considered because western California has a vast potential for solar electricity production and the Central Valley for hydropower which could be leveraged to provide electricity to the nearby industrial facilities. Specifically, there are 5 glass facilities that lie within the Central Valley and would have access to hydropower for their operations. Lastly, in the southernmost part of California, there is a hub of geothermal energy sources that can be used to provide power to the nearby lime facility.

4.1.5. Storage and transportation

Many of the above cited industrial facilities are located within 150 km of a sedimentary storage reservoir in the Central Valley, the most promising locations being the depleted oil and gas fields in the southern Sacramento and the southern San Joaquin basins [227]. While local in-depth field studies should be conducted to assess the pertinence of specific CO₂ storage sites, potential sites and CO₂ pipeline routes have been identified [228,229]. In addition to technical considerations, basins located close to population centers, such as the Los Angeles basin, are unlikely to be used for CO₂ storage due to safety concerns. Facilities located far from CO₂ injection sites in the Central Valley sedimentary reservoirs might have alternative storage opportunities in basalts and ultramafic rocks, as soon as commercial demonstration projects show the viability of these methods. The distance and the means of transportation between the CO₂ capture site and the CO₂ sequestration site is an important parameter to consider as longer transportation distances increase the overall costs and decrease the CO₂ avoidance potential. Facilities with capture volumes below 500 ktCO₂/yr and located close to CO₂ injection sites might favor truck transportation, while a joint access

pipeline connecting farther facilities with large capture volumes (> 500 ktCO₂/yr) to the storage location could be a more economically attractive option [65]. Also, most of the above cited industrial facilities are located close to the rail network that passes through the Central Valley. Rail could thus be considered for biomass transportation from processing facilities and for CO₂ transportation to storage sites.

4.2. Pennsylvania and the decarbonization of heavy industry

4.2.1. Context

In Pennsylvania, the total gross emissions amounted to 233.2 MtCO₂eq in 2017, 31% being from industrial sources [231]. The heavy industry in the state of Pennsylvania includes 7 cement, 3 lime, 7 glass, 15 steel facilities, resulting in 2.8 MtCO₂eq, 1.2 MtCO₂eq, 0.6 MtCO₂ed, and 4.9 MtCO₂eq emissions respectively [18]. These industrial facilities are located near or on top of the sedimentary reservoir that resides under the majority of the west and northern parts of the state (Fig. 15). With such close access to potential CO₂ storage sites, the facilities that do not expect to retire their kiln or furnaces within the next 10–20 years should consider adopting CCS, as transportation costs for the captured CO₂ could be minimal. If each of the facilities that are projected to retire after 2030 would implement point source capture, this could result in the capture of 2.2 MtCO₂/yr, 0.6 MtCO₂/yr, and 0.6 MtCO₂/yr from the cement, lime, and steel industries, respectively, totaling to 3.4 MtCO₂/yr (36% of emissions from the heavy industry, and 4.2% of the total in-state industrial emissions).

Local opportunities for fuel switching are limited as large sources of nut shells or fruit stones and gray or blue hydrogen are located out of the state, driving up the transportation costs. Currently, 5 cement facilities out of 7 are combusting coal in their kiln [18], which may be motivated by the large production of coal in the state. For facilities that are expected to retire or upgrade their technology, oxyfuel and electric kiln technology should be considered. Specifically, the four cement plants located in the eastern part of Pennsylvania that are all centered near an industrial air separation unit (ASU). A partnership could be forged between these facilities and the industrial ASU to readily provide oxygen for oxyfuel cement kilns. Another cement plant with a similar

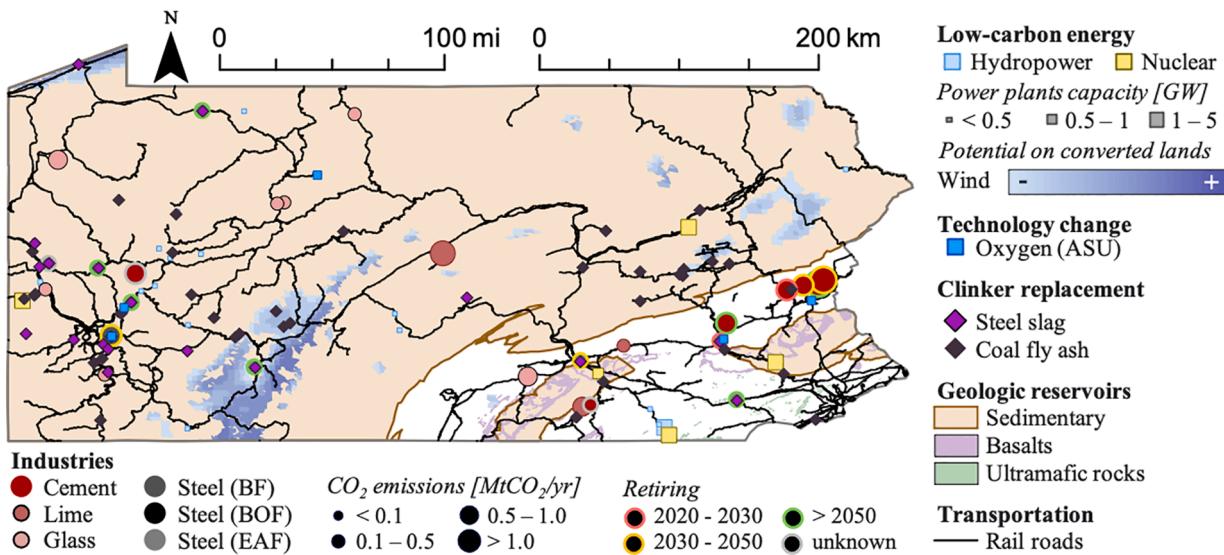


Fig. 15. Map of Pennsylvania highlighting cement, lime, glass, and steelmaking operations with estimated retiring year. These are overlaid with CO₂ storage opportunities, regions suitable for low-carbon energy, established oxygen production from commercial-scale air separation units (ASUs) and areas that produce materials suitable for clinker replacement [18,106–111,213–216,225,230]. The list of ASU locations is only indicative, as this data is proprietary.

opportunity can be seen slightly more west than the hub of four, but it is not expected to retire its kiln until after 2050.

Electric kiln technology is also an option in Pennsylvania because of the low-carbon electricity that is available. Throughout the state, but specifically near larger pockets of population density (outside of Philadelphia and Pittsburgh), nuclear power plants provide low-carbon electricity, and opportunities exist for the development of wind energy (Fig. 15). The steel industry in Pennsylvania is already largely electrified with 14 EAF for 1 BF/BOF [18,107]. The BF/BOF located near Pittsburgh could also transition to an EAF. However, EAFs tend to produce lower quality steel than the BF/BOF process. The total electrification of the steel sector should be considered carefully to avoid the disruption of supply for industries requiring high grade steel. It may be necessary to keep a minimum number of BF/BOFs in operation to ensure higher quality steel production for industries in which this is crucial.

The coal power plants and the steel industry are generating large amounts of fly ash and steel slag, which can replace part of the clinker in the concrete mix. The use of these materials can reduce the need for clinker for the construction industry and the GHG emissions associated with its production. Large amounts of fly ash and steel slag are produced, about 900 ktonne/yr and 600 ktonne/yr, respectively [230]. In addition, the byproducts stockpiled during the past years of operation could also be used for clinker replacement. Most of the locations with fly ash and steel slag are connected to the rail network, which can help transport these materials across the state.

5. Conclusions

Decarbonization of the cement, lime, glass, and iron and steel industries is met with unique challenges due to the CO₂ emissions that originate from the chemical reactions of feedstocks and the high temperatures required for processing. Existing kiln and furnace technologies have been optimized for their specific processes. So, changes to the geometry, design, or fuel handling can come with tradeoffs. The timeline for decarbonization in these industrial sectors may be limited by the lifetime of existing kiln or furnace technology, but it could be accelerated with the adoption of energy and materials savings practices, CCS, fuel switching with biomass, or feedstock switching. A summary of the primary decarbonization options investigated here, and the current stage of development for each kiln, can be seen in Table 5. Some of these options will be further limited by the regional resources, such as geological storage options for CCS or the availability of waste biomass. Through the evaluation of regional resources, the pathways of innovation that are most applicable can be prioritized.

Facilities with kilns or furnaces approaching their end of life are in a position to replace existing technology with low-carbon infrastructure. A regional resource analysis can be used to determine the most appropriate pathways of innovation. For example, established technologies such as fluidized bed or electric kiln technology coupled with CCS could be used at facilities with lower material throughput and access to low-carbon electricity. On the other hand, for a facility located near an oxygen producer, an oxyfuel kiln with CCS could be a viable option. Further and more in-depth analyses can be conducted to better highlight

Table 5

Summary of development stages and applicability of decarbonization solutions for each industrial kiln.

	CCS	Biomass Fuel Switching	H2 Fuel Switching	Fluidized Bed Kiln	Oxyfuel Kiln	Electric Kiln
Cement Kiln	Demonstration	Commercial	Research	Demonstration	Demonstration	Commercial
Lime Rotary Kiln	Demonstration	Demonstration	Research	Commercial	Research	Research
Lime Shaft Kiln	Demonstration	Demonstration	Research	Commercial	Research	Research
Glass Furnace	Research	N/A	Research	N/A	Commercial	Research
BF	Pre-Research	Research	Commercial*	Commercial**	Research	Demonstration***
BOF	N/A	N/A	Commercial*	Commercial**	Commercial	N/A
EAF	N/A	N/A	N/A	N/A	N/A	Commercial

* BF/BOF replaced with DRI process.

** BF/BOF replaced with FINMET or FINEX processes.

*** Molten Oxide Electrolysis pathway demonstrated by Boston Metal.

innovation pathways and potential partnerships that would be preferential.

Further development is needed to validate the emerging kiln technologies at a commercial scale, specifically hydrogen switching for natural gas in rotary and shaft kilns and electric kiln technology at high material throughput. Mapping studies should be conducted to determine the regional resources co-located with facilities to assist in developing technological roadmaps for the phase-out of carbon-intensive technology. Additionally, economic incentives for decarbonization in these industrial sectors should be considered to assist in offsetting additional costs of upgrading to low-carbon kiln technologies.

Declaration of Competing Interest

No Conflicts of Interest have arisen in the preparation or publication of this article.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.pes.2021.100982](https://doi.org/10.1016/j.pes.2021.100982).

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