this is less likely a problem with the metal-on-plastic strategy, given that PET's tensile strength is 170 MPa, comparable with Cu (210 MPa) and higher than Al (90 MPa). Finally, although lighter current collectors could decrease the raw materials cost of lithium-based batteries, more advanced processing techniques with increased costs remain as the main barrier to the effective integration of these developments to industrial manufacturing.

CONCLUSION

As electrode and electrolyte materials in high-specific-energy lithium metal batteries have become optimized, the percentage of "dead weight" has increased. These changes have made current collector weight reduction an attractive opportunity to increase the specific energy of cells by 10%–20%. Two representative lithium battery systems were analyzed to quantitatively understand the accessible gains through making current collectors lighter, demonstrating how, by optimizing one component, the requirements to meet long sought-after goals can be substantially eased in other areas of more challenging development. Multiple attempts at optimized and multi-functional current collectors have been recently developed. Although challenges remain, particularly with regards to scalable production, the potential and incentives for improvement are clear, and advanced current collectors will be a key component in batteries for clean energy storage and numerous other future-oriented applications.

SUPPLEMENTAL INFORMATION

Supplemental Information can be found online at https://doi.org/10.1016/j.joule.2021.03.027.

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Commentary

Decarbonizing cement production

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Paul Fennell is a professor of clean energy at the Department of Chemical Engineering, Imperial College London. His work encompasses the decarbonization and re-thinking of industrial processes, including the production of iron and steel and cement manufacture. He also works in the broader field of industrial decarbonization, including synergies between industry and power generation.

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emissions in order to mitigate climate change effects.

Introduction

The global production of ordinary Portland cement (OPC) is approximately 3.5 billion tons annually. It is a crucial material for building, used to make concrete, mortar, and other products. However, increasing attention is being paid to cement-related CO_2 emissions, which are significant—constituting around 7%[1] of total annual energy and industry emissions. Environmental and annual reports of major Western companies (Cemex, Heidelberg Cement, and LafargeHolcim) reveal that 561–622 kg of CO_2 is emitted per ton of cement produced, with differences related to the materials used to produce the cement, the type of cement kiln used, and the fuels being burned, though there can be significant variation globally.

OPC is composed of a number of materials, the most important and largest proportion (around 95 wt %) of which is "clinker" and supplementary cementitious materials (SCMs) (discussed below). The remaining 5 wt % is gypsum (added to aid in controlling setting time). Figure 1A shows the current process for production of cement, which includes three main stages: raw material extraction and preparation, clinker production, and cement grinding. Limestone ($CaCO_3$) is the main feed constituent. Our baseline is a modern cement plant, fired exclusively with fossil fuels.

After the limestone is ground together with other minor constituents, the raw material is calcined at 900°C through a series of cyclones. The majority of the energy needed and the CO_2 emissions are products of the calcination process; the pre-calciner uses around 60% of the total energy and produces unavoidable "process" emissions, around 60% of the total CO_2 from the cement plant.[2,3] Once the mixture leaves, it enters the rotary kiln, where reactions occur at 1,450°C–1,500°C, and cement clinker (a mixture of calcium silicates) is produced. Concrete is produced when aggregates are added to the cement. The main reactions during setting involve the (rapid, over the course of hours to days) hydration of calcium trisilicate (Ca_3SiO_5) alite and (slower, over days to weeks) hydration of Ca_2SiO_4, belite, to cause bulk hardening via precipitation of calcium silicate hydrates and calcium hydroxide. Both reactions continue until completion.

The aim of this commentary is to give a brief and very simple overview of the main decarbonization options and their interplay in terms of direct emissions from the cement production process, approximately bounding their relative importance and value.

Technologies to reduce CO_2 from cement production

Here, we discuss the potential for decarbonization of the cement production process via different measures, as well as their combinations, as shown in Figure 1B. The capital costs will not be explored; rather, we focus only on the potential, challenges, and ultimate limits of different technologies.

A modern plant requires ~3.3 GJ of thermal energy per ton of clinker,[2] around twice the thermodynamic minimum (the energy needed to calcine the limestone and to drive the clinkering reactions). Efficiency improvement has been incremental in recent years, as expected in a mature industry; the average thermal intensity across the globe fell from 3.75 GJ/t for clinker in 2000 to 3.5GJ/t in 2014, around −0.5% a year.[4] Given that both process and fuel-related emissions account for a significant proportion of the total direct emissions, both process modification and energy efficiency are important for CO_2 mitigation. Energy efficiency can be improved through energy recovery, waste heat recovery, and increasing the proportion of dry and semi-dry processes[2] (essentially, historically plants would feed limestone to the kiln by producing a slurry using water, but the evaporation of this water is very energy intensive). Carbon capture and storage (CCS) has significant potential, with numerous pilot and larger-scale demonstrations planned. It is also possible to use alternative fuels (refuse-derived wastes, biomass, or hydrogen) in the kiln or potentially to electrically heat parts of the process. However, it must be considered that in some cases these alternatives could reduce direct emissions but increase indirect emissions, in particular for hydrogen and electrification. Finally, emissions might also be reduced through reducing demand for clinker, such as by substituting alternative materials. Figure 1B outlines these major approaches to mitigating CO_2 emissions from cement production, discussed further below.

Waste heat recovery (WHR) (assumed that minor improvements continue, allowing 5% improvement in fuel use)

There are significant heat losses (about 35%–40%) from a cement manufacturing plant, mainly as a result of the air stream used to cool the clinker to 100°C and to the flue gases. A significant fraction of the heat input to the system is lost by convection from the pre-heaters and kiln, clinker discharge, dust, and radiation. We have not considered electricity demand in our analysis; it is assumed that the sum total of electricity production via WHR can offset plant electricity demand, and this might require both a traditional steam WHR system and an organic Rankine cycle. Both steam-based and organic Rankine-cycle-based systems can be used to convert waste heat to electricity, and this can offset a large proportion of the electricity demand of a plant.[5] The analysis below only considers direct CO_2 emissions from the process and fuel use.

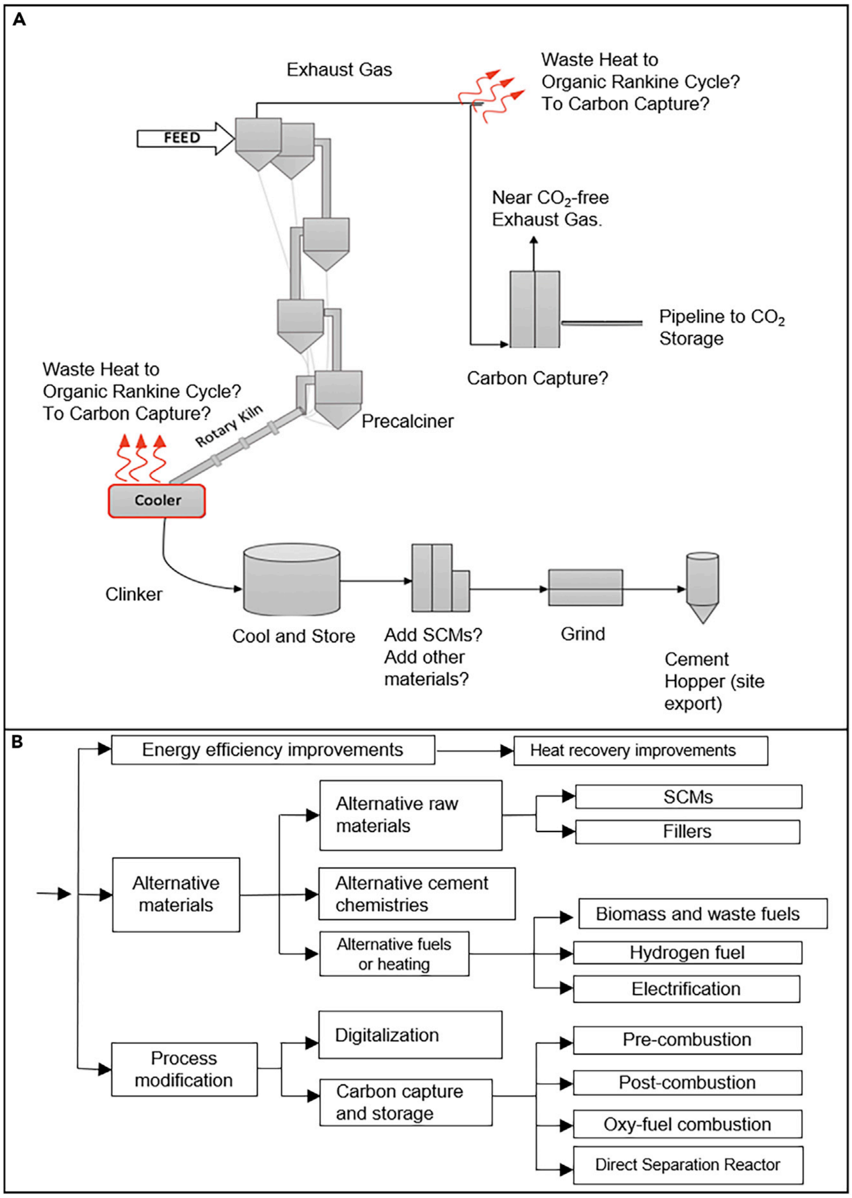

Figure 1. Cement production and decarbonization

(A) Cement production process. Stages with a question mark are not core to the process.

(B) Potential methods to reduce the CO_2 emissions from cement production.

Lower-clinker and lower-carbon cements (baseline assumption 0.7)

SCMs are materials that have cementitious properties and therefore can be used to replace cement clinker.[6] Many materials have been tested as SCMs, from copper tailings to sugar cane bagasse to the more commercially tested (but possibly reducing in availability as the world moves to net zero) pulverized coal fly ash (PFA) and ground granulated blast furnace slag (GGBS). Of course, some SCMs have emissions associated with their production, and the partitioning of these emissions between their production and final use is complex. Here, we focus on direct emissions. Given that construction managers have become more comfortable with these substitutions because of greater experience globally with them, the amount of clinker in a given volume of cement has decreased. Currently, the average global clinker ratio (kg of clinker per kg cement) is around 0.7, but this is heavily influenced by Chinese clinkers, which are now starting to use fewer SCMs.[7] We assume a base clinker content of 0.7, a reasonable reduction to 0.6 using current SCMs, but have investigated the effects of clinker down to a ratio of 0.5, which might be appropriate for so-called LC^3 (limestone calcined clay cements). These are a promising type of cement[8] that is similar to currently commercial cements and so might face lower barriers to commercialization than other novel cement formulations.

Alternative fuels (assumed substitution for MSW [municipal solid waste] up to 100%, biogenic fraction 0.7)

The use of alternative fuels can substantially reduce overall emissions, compared with those of a fossil-fueled plant, but ensuring the climate benefits requires careful analysis to traceably certify such reductions. According to the International Energy Agency,[9] between 60% and 80% of the carbon in municipal solid waste is biogenic in nature; a biogenic fraction of 0.7 has been assumed here. The replacement of fossil fuels with waste-derived alternative fuels is a cost-saving way to reduce fossil fuel use, in addition to being a relatively environmentally friendly method of waste management, particularly if care is taken to divert all recyclable material prior to use in the kiln. Biomass fuels are another option for reducing emissions, assuming that the biomass is CO_2 neutral. Lafarge, as part of the Canadian Cement 2020 project, has run up to 10% substitution at its Bath cement plant in Ontario. The fuels used were hemp, sorghum, willow, switchgrass, and oat hulls in the first phase, with more challenging fuels such as demolition wood, treated telegraph poles, etc. (and investigation of pre-processing of fuels to enable a hotter flame) in the second phase. Reports were positive, and permits were applied for to increase substitution up to 30%, though little has been reported since 2018. However, recent reports suggest that biofuels (outside of wastes) remain several times more expensive per unit of heat provided than waste fuel or coal.[10]

In a similar vein, hydrogen fuel and/or electrification could potentially provide up to 100% of the heat in the system, but compared with the magnitude of reductions in CO_2 emissions, the complexity and cost of designing and deploying hydrogen-driven kilns are high.

Importantly, no fuel avoids "process" emissions from calcination of $CaCO_3$ (~60% of the total).

Digitalization (assumed possible improvement 10% of fuel burned)

The benefits to cement plants from improved process control and next-generation measurement devices are significant. LafargeHolcim is a leader in this area and has started a "Plants of Tomorrow" initiative, which will roll out technologies including robotics, AI, and predictive maintenance.[11] A plant certified under the scheme is stated to have a 15%–20% operational efficiency gain. More than 80% of LafargeHolcim's cement plants are already connected to its technical information system, allowing performance tracking and allocation of resources centrally.[11] Given the challenge in converting a company's stated operational efficiency gain to decarbonization, an approximate value of 10% has been assumed.

Carbon capture and storage (assumed that 100% CO_2 capture is possible)

Traditionally, emissions from both the kiln, and calcining limestone, are combined into a single-flue gas stream that passes through the preheating train and out of the plant. Because of the high CO_2 concentration (14%–33% by volume), basic thermodynamics indicate that it is in general easier to capture CO_2 from a cement plant than an equivalently sized natural gas (~3%) or coal-fired (~15%) plant.[12] Of course, the presence of dust, NO_x, SO_2 and SO_3, and trace and minor species might complicate this simple picture. There are in general three different types of CCS: post-combustion capture, oxyfuel combustion, and pre-combustion (here covered under "hydrogen fuel"). In the cement context, the direct separation reactor (DSR) has been recently developed and is in the process of commercialization by the Australian company Calix.[13]

Post-combustion capture involves removing CO_2 from flue gases. It is either retrofitted into existing plants or built as an end-of-pipe capture technology for new plants. It is not necessary to go into detail as to the different types of post-combustion capture except to mention that there are multiple types, with some more suited to cement production than others. There might be an increased energy demand for certain types of CCS.

In oxyfuel combustion, capture of CO_2 takes place after the fuel is burnt with pure oxygen (and recycled CO_2) instead of air. It was the first technology heavily promoted for CCS on cement, because removing the large proportion of nitrogen in the gas flowing through the plant theoretically lowers fuel consumption. There are no significant issues with cement produced under an oxyfuel atmosphere;[14] however, it is difficult to retrofit the process to an existing cement plant. This type of CCS allows 100% CO_2 capture. Capital costs for the oxyfuel plant might mean co-location with an oxygen producer, possibly as part of an industrial complex, could be preferable.

The DSR comprises a novel method to calcine limestone. A large, externally heated tube calcines ground limestone as it falls through it vertically. This re-engineering of the process allows the capture of the almost-pure CO_2 released from the limestone without substantially increased energy use (except a potential decrease due to less efficient indirect firing in the calciner) or significant additional costs.

It is noteworthy that Norcem's Longship CCS project in Brevik, Norway, has recently received a positive final investment decision by the Norwegian government for a full-scale demonstration, leading to a greater degree of certainty that CCS will be available for cement.[15]

Results

This section estimates the net CO_2 emissions in relation to a baseline of a cement plant (only direct emissions, i.e., those from fuel and calcination are considered) with a fraction of clinker, (S) of S_{base} (0.7, as discussed above), with no process efficiency improvements and utilizing exclusively fossil fuel. Of course, these estimations are not precise and need proper life-cycle analysis to be validated, particularly in the case of bioenergy and hydrogen production. Additionally, they do not take into account efficiency changes as the fuel to the kiln is changed or changes in the fuel C/H ratio between coal and biomass but are a first approximation of trends in emissions.

The total CO_2 emissions can be calculated with Equation 1, where $CO_{2(process)}$ is the process CO_2 emissions in relation to the baseline cement (before considering carbon capture), $CO_{2(fuel)}$ is the fuel-related CO_2 emissions and is calculated with Equation 3, $CO_{2(biomass)}$ is CO_2 taken up during the growth of biomass, estimated by Equation 4, and $CO_{2(captured)}$ is CO_2 captured by CCS technology, calculated with Equation 5. $CO_{2(reabsorbed)}$ is CO_2 reabsorbed through the process of natural carbonation.

$$CO_{2(total)} = CO_{2(process)} + CO_{2(fuel)} - CO_{2(biomass)} - CO_{2(captured)} - CO_{2(reabsorbed)} \quad \text{(Equation 1)}$$

Here, $CO_{2(process)}$ is estimated by Equation 2, with the fraction of process emissions, P approximately 0.6, as discussed above:[2]

$$CO_{2(process)} = \frac{S}{S_{base}} P. \quad \text{(Equation 2)}$$

We have estimated CO_2 emissions from the fuel in Equation 3 by considering D (the fractional decrease in emissions due to digitalization, which we have set to 0.1 where it is considered), $(1 - P)$ is the fraction of fuel emissions, E is any efficiency gain in the process (0.05 where considered), and H is the fraction of hydrogen (by energy content, assuming no change to efficiency by switching to hydrogen). Results for direct emissions from fuel switching to hydrogen are equivalent to those from electrification.

$$CO_{2(fuel)} = \frac{S}{S_{base}} (1 - D)(1 - E)(1 - P)(1 - H) \quad \text{(Equation 3)}$$

To determine equivalent emissions offset by the use of a biogenic fraction of the fuel, F, which is assumed to be CO_2 neutral, we have

$$CO_{2(biomass)} = \frac{S}{S_{base}} (1 - D)(1 - E)(1 - P)F, \quad \text{(Equation 4)}$$

and where C is the fraction of CO_2 capture, the total CO_2 captured is given by

$$CO_{2(captured)} = C\left(CO_{2(fuel)} + CO_{2(process)}\right) \quad \text{(Equation 5)}$$

Finally, once concrete is put in place, it reabsorbs CO_2 through the process of natural carbonation. This process takes place over a decadal time span; thus, it is usually not included. However, for the purpose of completion, it was considered in Equation 1. According to Xi et al.,[16] the fraction of $CO_{2(reabsorbed)}$, R, is around 5% of the total CO_2 emitted during current cement production, though this can vary substantially depending on a host of factors.

Figure 2 shows the effects of the application of CSS for a variety of fossil- and non-fossil-based fuels, with the effects of efficiency improvement, clinker substitution, and their combination shown for each fuel.

Discussion and conclusions

The "zero" CCS case, for a fossil-fuel-fired cement plant is instructive to examine first (i.e., the left-hand axis of Figure 2A). Changing the clinker ratio or plant efficiency, though highly financially profitable, make small improvements in overall CO_2 emissions, though in combination they have a noticeable effect. It is clear that a shift to, e.g., LC^3 cement would have a more significant effect and also that CCS would be necessary for near net zero emissions.

Figures 2B and 2D show that changing the fuel to hydrogen (or using electricity or biomass) without CCS does not do anything for process emissions. It is for this reason that altering the kiln efficiency does not change the direct emissions—fuel emissions are zero, and what is shown is, in effect, simply the process emissions. Furthermore, for all cases, clinker substitution has a more significant effect on emissions than improved process efficiency, given that clinker substitution also reduces the process emissions.

There is a clear diversion in emissions between hydrogen and biomass as a fuel as CO_2 is captured, with a biomass-fired kiln becoming CO_2 neutral at around 63% CO_2 capture, MSW at around 80% and the other fuels only at (the near impossible) 100%. Of course, combining all technologies (other than CCS) leads to a significant reduction in CO_2 emissions, but even combining all technologies (except LC^3 cement) and firing the kiln with either biomass or hydrogen leads to 50% of the base-case emissions, far above those required for net zero.

As expected, for low or zero deployment of CCS, increased efficiency or clinker substitution reduces the overall emissions per ton of cement. However, for very high CCS deployment with biomass or MSW, the (counterintuitive) opposite is the case—because burning more biomass per ton of cement produced (i.e., being less efficient or having a lower clinker substitution) results in greater negative emissions of CO_2. It is important to note that biomass will be a limited resource and that the main value from the process is in the cement produced and not the negative emissions (otherwise, one would not make any cement, just burn the biomass and sequester the emissions directly). It is also important that although this simple analysis can give insights into what is and is not critical for net-zero or negative emissions, a full and comprehensive life-cycle analysis is necessary in order to fully quantify emissions.

Only one point has been shown for "reabsorption," but reabsorption of CO_2 would shift all points downward by around 0.05 (it could be argued that the baseline should also include some reabsorption). This has the effect of potentially allowing CO_2 neutrality at only 90% CO_2 capture with fuels other than biomass.

The most striking feature of this simple analysis is that it is the underlying biogenic component of the fuel, together with the addition of CCS, that makes the biggest reductions in CO_2 emissions possible. Clinker substitution is valuable, particularly when CCS is not deployed, but very deep decarbonization will require going further. Given that MSW is very much less expensive than biomass as a fuel[8] but

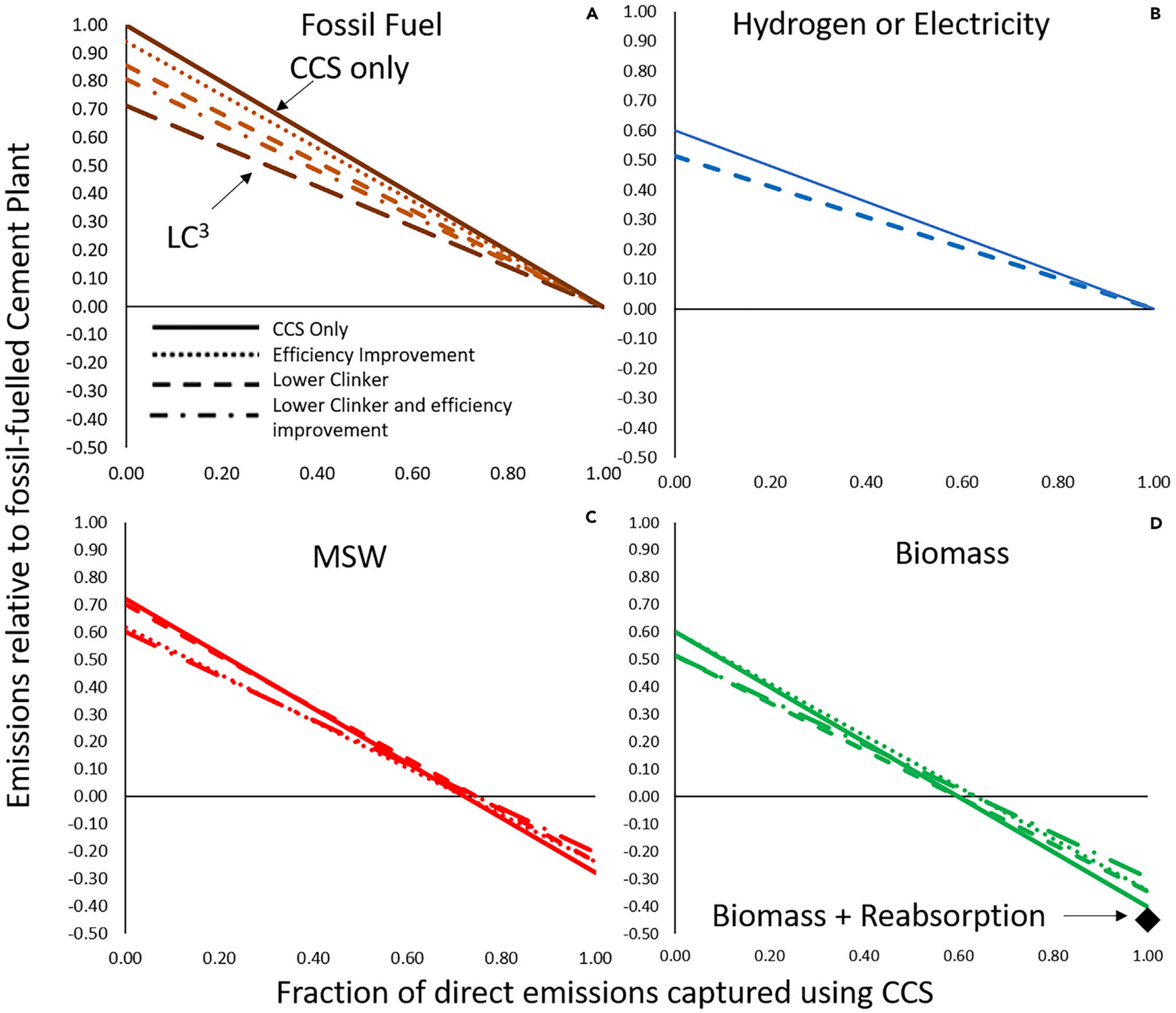

Figure 2. Relative CO_2 emissions for different combinations of fuel with CCS

(A) Fossil fuel only.

(B) Hydrogen or electrification.

(C) MSW (Biogenic fraction = 0.7).

(D) Biomass (biogenic fraction = 1).

Shown also for each plot are the effects for each fuel of changing to lower clinker ----, improving the efficiency of the process ●●●, and their combination ●-●-. Baseline clinker factor 0.7 (in all cases lower clinker = 0.6, LC^3 = 0.5 where shown), efficiency improvement in all cases = 0.10 via digitalization, 0.05 from energy efficiency. Fraction of process emissions = 0.6. Reabsorption fraction when shown = 0.05.

adequate in very high proportion for cement production, this indicates that for a net-zero world, the deployment of CCS is key but that cement could potentially produce significant quantities of "negative emissions" with an MSW-based fuel, for which there will be significantly less demand than clean biomass.

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AUTHOR CONTRIBUTIONS

Conceptualization, P.S.F. and S.J.D..; methodology, P.S.F.; investigation, P.S.F. and A.M.; writing – original draft, P.S.F. and A.M.; writing – review & editing, S.J.D.; supervision, P.S.F.

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