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Techno-economic assessment of calcium sulfoaluminate clinker production using elemental sulfur as raw material

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ABSTRACT

The foreseen decarbonisation of the cement industry by 2050 will require a combination of different measures in the current manufacturing practices such as the improvement of the energy efficiency, the use of alternative fuels, the reduction of the clinker to cement ratio, and the integration of carbon capture, especially beyond the year 2030. The production of alternative binders, for the substitution of the current product families of cement, is under investigation due to its significant potential in emissions reduction, although robust and agreed life cycle assessment methodologies are still claimed by the industry in this field. Calcium sulfoaluminate (CSA) cements appear as a promising alternative to Portland cements due to the significant reduction of the direct emissions of carbon dioxide caused by the lower need of limestone and fuel. However, they require sulfate and higher inputs of aluminium oxide, sourced from bauxite. This study focuses on the techno-economic performance, in the geographical scope of Qatar, of a novel CSA production method consisting of the combustion of sulfur to supply SO_3/SO_2 to the clinkering reaction. This new approach is proven to reduce the associated greenhouse gases emissions of Portland cement clinker production, while, in general, CSA clinker production costs are higher than those for Portland cement clinker. Nevertheless, CSA has a competitive CO_2 avoidance cost, provided that the economics of raw materials supply, such as bauxite, favours its production; an aspect that is further emphasised in a prospective analysis towards more restrictive climate policies.

1. Introduction

The cement industry is responsible for the production of around 5–8% of greenhouse gases (GHG) emissions and of 27% of industrial emissions worldwide (Kajaste and Hurme, 2016; van Ruijven et al., 2016). However, it is expected that, especially due to the rising global population and urbanisation patterns, the global production will increase by 12–23% by 2050 from current levels of production, increasing by 4% the direct emissions from the sector (Cement Sustainability Initiative and International Energy Agency, 2019).

In such scenario, the cement industry has made a commitment for the decarbonisation of their activities beyond the 2 °C increase scenario, where a combination of different measures in the industry would be able to reduce by 24% the greenhouse gases emissions of the sector in 2050 from current levels. Those technological measures are: improving energy efficiency, using of alternative fuels, reducing the clinker to cement ratio and integrating carbon capture, especially beyond the year 2030 (Cement Sustainability Initiative and International Energy Agency, 2019).

Another technological measure is the production and use of alternative binders, which can substitute the use of current families of cement. Gartner and Sui (2018), described different families of binders to be considered in the medium-term as low carbon alternative formulations: reactive belite-rich Portland clinkers, belite – ye'elimite-ferrite binders (BYF), carbonatable calcium silicate cement, and magnesium oxides derived from magnesium silicates. Although there are possibly more alternatives, these are the new binders that the industry could adopt without major changes of current practices, and are the more likely to become commercial in the near future market (Gartner and Sui, 2018). However, the potential environmental benefit from its implementation has not been taken into account in the decarbonisation routes for the cement industry due to the uncertainty associated to the

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deployment of alternative binders deployment and their impact quantification.

In this context, BYF cements, which in this paper are referred to as calcium sulfoaluminate cements, CSA, are proposed as a low carbon footprint alternative to Portland cement that would not require a technological change in the cement industry; a Portland cement kiln could shift easily its production to CSA. A recent calculation showed an achievable reduction of 35% of greenhouse gases emissions (Theodore Hanein et al., 2018a), although the eco-efficiency of its production is restricted by many factors associated with the supply chain (Gálvez-Martos et al., 2020).

CSA cements are mainly composed of belite, ye'elimite and ferrite. They consume less energy than conventional Portland cement, since the production of alite is avoided, which requires higher clinkering temperatures (over 1400 °C), and produce ye'elimite, which is the mineralogical name for CSA, with the formula $Ca_4(AlO_2)_6SO_3$ and able to form ettringite during hydration, an important phase in cement strength development (Zajac et al., 2018). Ye'elimite introduces two important differences in the formulation: a high content of aluminium oxide and sulfate as new main ingredients in the composition of clinker. The extra requirement of aluminium oxide is usually sourced from bauxite or bauxite-mining wastes, which increases production costs.

As source of sulfate, gypsum is usually the preferred choice in existing commercial CSA formulations. In this context, the Qatar based Gulf Organisation for Research and Development (GORD) proposed the use of sulfur (Al Horr et al., 2017), supplied from desulfurization operations in oil refineries and natural gas treatment, which is available in Qatar; sulfur would act as a fuel (Hanein et al., 2016), reducing further the carbon footprint of the process (Theodore Hanein et al., 2018a), and would provide enough SO_2/SO_3 in the formation of CSA.

The balance of CO_2 of novel CSA formulations against Portland cement clinker is proven remarkably favourable in terms of direct emissions from cement making (Habert et al., 2020) from a life cycle perspective (Ren et al., 2017) and more aggressive policies in terms of industrial decarbonisation could result in a faster deployment of low-carbon technologies and options, such as CSA production and such as sulfur-burning approaches. The main factor negatively affecting the economics and the carbon footprint of CSA production is the use of alumina sources, which require bauxite or bauxite-derived raw materials, available only in a few parts of the world. This introduces important drawbacks in the potential deployment and exploitation of CSA formulations; research has then been oriented to develop CSA formulations at a low content of alumina (low ye'elimite), which can alleviate the impact of the alumina source on the environmental and economic performance of CSA making (Elhoweris et al., 2018).

A recent paper evaluated the eco-efficiency of CSA clinker production, and concluded that it is a locally oriented solution for alternative sustainable cement making (Gálvez-Martos et al., 2020), i.e. it is only eco-efficient in those locations with availability of bauxite or aluminium oxide useable minerals (e.g. as high alumina clays) or within a certain range of aluminium oxide sources. Only in those locations, eco-efficient CSA product systems are found with similar costs to Portland Cement clinkers and a significantly lower carbon footprint.

However, the Middle East has been reported as having a large cement production capacity due to its souring construction activity (Hasan and Karmani, 2013). In this context, the strategic geographical situation of Qatar, with a high aluminium production capacity and therefore with a stable and voluminous trade of bauxite, can make the country a business case for CSA production. In addition, there is a high availability of sulfur due to the oil and gas industry, and a high demand of cement for the construction sector in the country: the apparent consumption of cement is above 3 tonnes per capita, while in the U.S. it is below 300 kg per capita (Globbulk, 2020). All these factors, along with a political aim for the use of locally available resources could make CSA cement to play a key role in Qatari cement market.

This study aims at the analysis of the techno-economic performance of CSA making in the context of Qatar using sulfur combustion, focusing on (i) the potential geographical origin of the bauxite source, (ii) the fuel used, (iii) the methodological approach of considering sulfur as a waste with zero environmental burden, and (iv) the need for a wet scrubbing system for extra SO_2 emissions. For this last point, the cost balance was further analysed in a prospective scenario to 2050, taking into account the expectations for the cement sector decarbonisation and the cost of CO_2 emissions in trading schemes.

2. Materials and methods

2.1. Steps of the analysis

The techno-economic analysis presented in this work is divided in the following stages:

- In the first stage, the mass and energy balance of a selected group of formulations was calculated; the input-output balance was used to estimate the carbon footprint and the potential economic performance of the targeted formulations of CSA clinkers and the reference Portland cement clinker.
- In the second stage, the analysis was extended using a sensitivity analysis of the fuel used in the kiln, the methodological approach for sulfur as a waste or as a priced commodity, and the geographical location of the bauxite source.
- In the third stage, a prospective analysis of the cost of CSA was performed under the future market of CO_2 emissions and under the scenario of cement decarbonisation provided by the technology roadmap of the Cement Sustainability Initiative (Cement Sustainability Initiative and International Energy Agency, 2019).

2.2. Analysis approach

The main indicator used in this assessment is the CO_2 avoidance costs, which was considered an appropriate indicator for the techno-economic benchmarking of low carbon alternatives. This is defined as indicated by the following equation:

$$Avoidance\ cost = \frac{Cost\ of\ low\ carbon\ option - Cost\ of\ conventional\ option}{Emissions\ from\ low\ carbon\ option - Emissions\ from\ conventional\ option}$$

This indicator is commonly used in carbon capture and storage (CCS) or utilization (CCU) assessment, since it gives a value to each avoided unit of impact. In the context of power generation with CCS processes, it only accounts for direct emissions, since it can be used by plant owners and investors to assess CCS performance against associated cost in emissions trading schemes or emissions-related taxes (Simbeck and Beecy, 2011). The use of life-cycle associated emissions would always be required when assessing low carbon options that involve a significant change in the supply chain of raw materials or energy, as it is the case of alternative cementitious binders (Theodore Hanein et al., 2018a); however, regarding alternative binders, the industry still lacks of "*considerable further analysis [...] to produce robust, independent and publicly available life cycle assessment of these materials, including a comparative quantification of the production costs and their long-term performance*" (Cement Sustainability Initiative and International Energy Agency, 2019). These considerations made the industry plan their future climate mitigation commitments without taking into consideration the potential benefits of alternative binders. Only proven technologies towards the reduction of direct emissions and oriented to energy efficiency, carbon capture, reduction of the clinker-to-cement ratio and the use of alternative fuels were preferred.

Alternative binders are excluded from the decarbonisation roadmap of the cement industry. This is probably a consequence of the current emissions trading schemes or carbon tax mechanisms, introduced voluntary by countries after the Kyoto protocol. For example, the EU emissions trading scheme (EU ETS) does not include indirect emissions, not even those derived from the consumption of electricity (European Commission, 2015). Since alternative binders, such as CSA, are related to the reduction of emissions in a life-cycle perspective, these require agreed robust methodologies in a larger scale, probably to come under the implementation of the Paris Agreement in a long term strategy (Government of Japan, 2019; OECD, 2018).

In CSA making, direct emissions from the kiln are reduced, but there is a well proven increase related to the supply chain of raw materials. So, for this work, the CO_2 avoidance cost was defined by the difference between the cost of CSA clinker and the cost of Portland cement clinker, divided by the difference of carbon footprint of the clinkers. However, the carbon footprint is preferred over direct emissions as a metric. It is unconventional to include indirect emissions such as those associated to the production of raw materials and fuels. But, in the case of CSA, indirect emissions are an extremely important factor in order to understand the actual potential of the alternative binder, since raw materials such as bauxite have a significant influence in life-cycle associated emissions. The life-cycle perspective introduced in this indicator brings out an important source of uncertainty in the comparison with literature data.

2.2.1. Costs estimation

In this work, the economic model developed by Barker et al. (2008), for the International Energy Agency, was adapted, mainly including costs from wet scrubbing and CO_2 were appropriate, and updated, using current commodity prices. The most important assumption in the economic model for CSA cements is that the process of CSA clinkering was assumed the same as for PC clinkering, so existing kilns can directly be used for CSA burning. The implication of such assumption is that no capital cost or major investment is required in the change of operation from Portland to CSA clinker production. However, there is a major concern in the use of sulfur as a fuel, which may be responsible for excessive SO_2 emissions and a wet scrubber would then be required.

The economic assessment includes variations only in operational expenses that depend on variable costs (i.e. raw materials and fuels), using the same structure for the fixed costs assumed by Barker et al. The actualised cost of Portland cement clinker is USD 60 per tonne of clinker, of which USD 42 are fixed costs and would be the same for 1 tonne of CSA clinker. This includes electricity (USD 6.6), maintenance (USD 15.6), labour (USD 7), administration (USD 1.8) and other fixed costs (USD 7.8).

2.2.2. Carbon footprint

The carbon footprint of the clinker formulation was calculated using a streamlined life cycle assessment (LCA) methodology. The approach was selected as a cradle-to-gate for clinker, i.e. grinding and additions for cement making were not considered, since they can be assumed equal for all the formulations. The method for the calculation of the global warming potential as carbon dioxide equivalent emissions was taken from the IPCC 2013 method (IPCC, Intergovernmental panel on climate change, 2013), using an attributional approach, where specific impacts are associated to specific processes. The product system boundaries includes all steps for cement clinker production: extraction, refinement, distribution of raw materials in the raw feed (known as raw *meal*), fuels, and the burning process to produce clinker (Fig. 1).

Since the system was defined in terms of clinker manufacturing, the selected functional unit is 1 tonne of clinker to compare and benchmark production systems. However, the results may not be comparable in terms of functionality of the final cement. Further discussion on this issue is provided in the discussion of the results.

Emissions data from background processes (e.g. such as those related to the supply chain of raw materials and fuels) have been generally taken from ecoinvent 3.6. Database unless otherwise stated in section 2.3. Life cycle impact assessment (LCIA) has been performed using the LCIA results provided by the ecoinvent database (Wernet et al., 2016) for background processes, using our own MATLAB-programmed user interface (see Supplementary Material).

2.3. Target formulations and supply chain

The primary difference between CSA and PC is the presence of ye'elimite in CSA. Ye'elimite, also known as Klein's salt or tetracalcium trialuminate sulfate, is an essential component of CSA-type clinkers. Novel CSA formulations are also known as BYF cements (belite, ye'elimite, ferrite, from its molecular composition). The order of the acronym BYF reflects the importance of each phase, being belite the most abundant phase, in contrast to more conventional CSA, where the clinker bears a 60–70% ye'elimite (Gartner and Sui, 2018). Ye-elimite and ferrite interact with calcium sulfate in the presence of water leading to the formation of ettringite and monosulfate during the hardening process. In this way, alite, the main component of Portland cement and which requires a high temperature for its formation, is avoided (Sahu et al., 1991), while still achieving good mechanical properties in cement-derived products.

Hence, CSA cements can reduce the associated emissions of greenhouse gases due to the less need of calcium carbonate, limestone, in the raw meal, and the less amount of energy required to achieve the final mineralogical composition. The targeted formulations were selected following the same approach as in previous studies (Gálvez-Martos et al., 2020), so the ye'elimite content of the final clinker was varied from 30 to 70%, ferrite was assumed at 10%, and the rest (20%–60%) was considered to be belite; the source of sulfate was elemental sulfur combustion. As a reference, 60% ye'elimite conventional CSA, made from gypsum, and conventional Portland cement clinker manufacture were also analysed.

As mentioned before, the manufacture of CSA cements requires of a substantially different supply chain due to the use of less

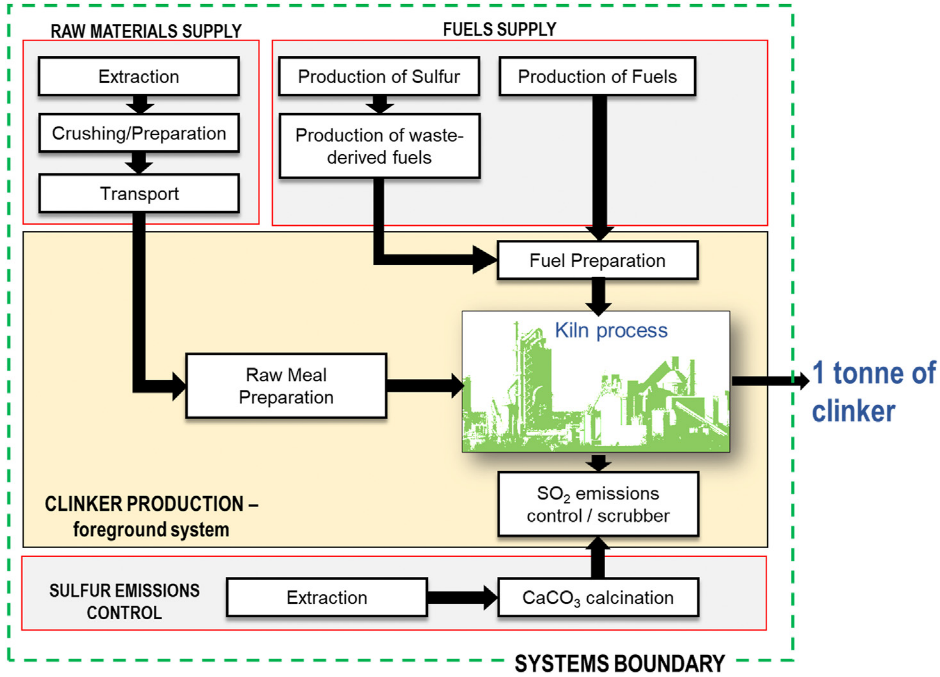

Fig. 1. Main flow diagram and systems boundary of the production system of 1 tonne of clinker.

Table 1
Transport distances from bauxite mines.

Origin	Road, km	Ocean freight, km
Guangxi, China	113	10,152
Gove, Australia	1022	10,002
Para, Brazil	579	16,879
Orissa, India	305	4895

limestone or fuels and the use of new components. The main assumptions for the supply of raw materials, fuels and sulfur are described in the following subsections.

2.3.1. Raw materials

Bauxite. Imports of aluminium oxide to Qatar come mainly from Australia (79%), Brazil (12%), and India (9%) (UN, 2018). For this work, China was also included as source of bauxite, since it is a common trader and has important reserves (USGS, 2018). For cost and carbon footprint calculation, the assumed transport distances by road and ocean freight are shown in Table 1.

The carbon footprint of bauxite mining is 6.2 kg of CO_2eq per tonne, according to the ecoinvent database (Wernet et al., 2016). Transport impact was calculated for each evaluated origin: road transport consumes 50 g of diesel per tonne kilometre (tkm), and 9 g of diesel per tkm by ocean transport. Diesel combustion emissions were assumed as 74 g of CO_2 per MJ, and the assumed low heating value, LHV, was 42.8 MJ/kg. The cost of bauxite mining is USD 31 per tonne f.a.s. (*free alongside ship*) (USGS, 2018), to which transportation costs were added, at USD 0.062 per tkm for road, and ocean freight at USD 0.0048 per tkm.

Clay. The assumed footprint for clay is 10.3 kg CO_2eq per tonne (Wernet et al., 2016), including the emissions associated to transport at 0.019 tkm per tonne of clay, contributing 24% of the total impact of clay. Its price was assumed at USD 13 per tonne, as per common clays, including transport, corresponding to the UN Comtrade reported data (UN, 2018).

Limestone and gypsum. These raw materials are locally available in Qatar (Trimmer and Szczesniak, 2019); limestone production adds up to 2.3 million tonnes, and gypsum sums 210,000 tonnes in Qatar in 2015. The average carbon footprint for them is 3.9 kg CO_2eq per tonne of limestone, and 3.8 kg of CO_2 per tonne of gypsum (Wernet et al., 2016). For transport, 80 km of road mode were assumed. Price is USD 22 per tonne of gypsum and around USD 3.5 per tonne of limestone, as assumed in Galvez-Martos et al. (2020).

2.3.2. Fuels

Natural gas is the main fuel used for clinker burning in Qatar. The carbon dioxide associated emissions from natural gas burning is 56 g per MJ. This is desulfurised gas, with an LHV of 47.1. MJ/kg (WBSCD, 2014).

Petcoke was considered in this work, although almost no kiln in the Middle East operates with that fuel. However, its study could be interesting from the point of view of CSA clinker production chemistry due to its high sulfur content (up to 4%) which makes it unusable in many applications but in cement kilns. Petcoke combustion, however, produces 93 g of CO_2 per MJ, with an LHV of 29.5 MJ/kg (WBSCD, 2014). Also, the cement industry worldwide uses petcoke as main fuel in many kilns, although it is increasingly being substituted by waste-derived fuels.

In the particular situation of the Middle East, caps to natural gas use have historically been implemented, which limited the production capacity of some countries and forced the production with alternative fuels imported from elsewhere (i3c, 2009). Even, there were kilns prepared for crude oil burning, which is more readily available than other fuels; however, the cost of such approach is unaffordable, and many kilns have already discarded that practice for the last 20 years. As for August 2020, the natural gas price in Qatar was USD 2 per GJ (Indexmundi, 2020), and the petcoke price in the Gulf Cooperation Countries (GCC) was USD 1.3 per GJ (Tridge, 2020).

2.3.3. Sulfur

In the proposed process alternative for CSA clinker burning (Al Horr et al., 2017), sulfur is fed as a supplementary fuel, which in the presence of oxygen is burnt to SO_2 and SO_3, which can react with calcium in the kiln to form ye'elimite, the characteristic component of CSA clinkers. The presence of an oxidising atmosphere containing SO_2 may have a beneficial effect on the chemistry of the clinker with better cementitious properties, e.g. as the stabilization of the α' belite polymorph with better reactive properties during hardening, as shown in the literature (Elhoweris et al., 2018).

Globally, elemental sulfur is produced by the Claus process during the desulfurization of natural gas (47%) and oil (53%), according to the ecoinvent database (Wernet et al., 2016). Sulfur is produced locally and is assumed to be transported an average of 80 km in Qatar.

Sulfur is commonly considered a by-product which is marketed and not a waste, so the LCA and economic balance assumed a carbon footprint of 241 kg CO_2eq per tonne of elemental sulfur, calculated from the inventory of the ecoinvent database with the IPCC method (100 years as time horizon). According to the UN Comtrade database (UN, 2018), a total of 402,000 tonnes of sulfur were exported in 2018 from Qatar at an average value of USD 136 per tonne, which was taken as a reference price for the use of sulfur in this study.

However, it was estimated that the production of sulfur is over 850,000 tonnes in 2015 (Trimmer and Szczesniak, 2019), so unmarketed stocks of sulfur may exist. Then, from an LCA methodological framework, it may be considered a waste, with a theoretical zero environmental burden and zero cost at its origin as cut-off criteria. This situation was considered and an alternative scenario for sulfur as a waste was analysed, where only the impact of sulfur transport was considered in the greenhouse gases balance.

2.4. Wet scrubbing

Experts acknowledge that low ye'elimite CSA clinker "*can be manufactured in standard Portland cement plants, which is a great advantage in terms of capital investment costs*." (Gartner and Sui, 2018); in fact, the shift to CSA making may require no significant additional investment, which would keep production costs similar to Portland cement if the ye'elimite content is low (Gálvez-Martos et al., 2020). But, the fact that the amount of sulfur, as SO_2 or SO_3, in the cement kiln increases significantly with respect to other conventional formulations may require a wet scrubber if the SO_2 emissions are over legal limits.

Sulfur dioxide emissions are a common problem faced by many conventional cement kilns. The Best Available Techniques (BAT) Reference Document for the Cement industry, published by the European Commission (European Commission, 2013), states that elevated sulfur dioxide emissions are expected when raw materials contain organic or readily oxidizable forms of sulfur (e.g. pyrites). This is not the case for high gypsum content, which is easily incorporated into the clinker. Also, the sulfur fed with the fuel to the kiln will condense as sulfate in the final product, since it is readily oxidised to SO_2 in the strong alkaline nature of the sintering zone and the calciner. However, the kiln, the preheaters and the calciner of modern kilns create sulfur cycles due to the reaction of SO_2 with calcium carbonate to form $CaSO_3$ and then $CaSO_4$ in the presence of O_2;when the solid reaches higher temperature zones of the kiln, the sulfate decomposes to CaO, SO_2 and O_2. For small amounts of sulfur in the feed, the rate of removal of sulfate in the solid is higher than the rate of removal as gaseous SO_2. If the input of sulfur in the raw meal is high, then the system has less capacity to remove sulfate and more gaseous SO_2 is released. The BAT document provides some indications on removal techniques for SO_2 as a function of their effectiveness and the total concentration of sulfur in the input: (i) optimisation techniques, such as varying the profile of temperatures in the preheaters or the increase of the amount of oxygen added at the low temperature end to avoid NO_x, (ii) addition of an SO_2 absorbent, such as hydrated lime, $Ca(OH)_2$, which capacity for retention is limited, and (iii) end of pipe treatments, such as wet scrubbing (only for SO_2 control) or active carbon filters, which are also indicated for other contaminants. For high sulfur input, oxygen concentration and absorbent additions are limited by the concentration and amount of SO_2. For higher flows of SO_2, wet scrubbing is the preferred technique in the cement industry; it consists of an end-of-pipe treatment of the gas with a slurry of calcium carbonate and oxygen, which transforms SO_2 into calcium sulfate and releases one molecule of CO_2 per molecule of reacted SO_2:

$$SO_2 + \frac{1}{2}O_2 + 2H_2O + CaCO_3 \rightarrow CaSO_4 \cdot 2H_2O + CO_2$$

Wet scrubbing has important implications in the techno-economic analysis of cement plants and, in the case of CSA making, it has a strong influence in the decision-making towards the shift of a PC kiln to CSA. It is expectable that the use of sulfur would require extra oxygen and, probably, some additions of absorbent to control emissions. The generated SO_2 is readily absorbable by the raw meal in the kiln (Hanein et al., 2016) and it is likely that it can be controlled without the need of wet scrubbing, given the current experience with high-sulfur fuels, as petcoke.

For this work, the economics of wet scrubbing were taken from those calculated In the BAT document for the cement industry (European Commission, 2013), which reports on the annualised cost (CAPEX plus OPEX over a 20 years time period) to be around EUR 1144 per tonne of removed SO_2, where operational expenses are EUR 460 (USD 494) per tonne of SO_2 (all costs actualised to 2019). For a standard kiln, with 1000 tonnes per day of production, the cost per removed ppm of SO_2 would be around EUR 1.4 10^{-3}. In terms of emissions, the CO_2 emissions increase by 1 mol of CO_2 per mol of SO_2 captured, i.e. 0.7 kg of CO_2 eq per kg of SO_2.

3. Results

3.1. Mass and energy balance

The mass and energy balances were calculated for the targeted formulations and are shown in Table 2. In the first column of the table, the label for each analysed scenario is given. For the CSA formulations, the case label defines the fuel used (NG for natural gas or PK for petcoke) and the following two digits show the targeted content of ye'elimite. REFCSA refers to the reference CSA clinker formulation, which is a commercial formulation of 60% ye'elimite made from gypsum, and REFPC refers to a reference case for Portland cement clinker from the literature (Chen et al., 2010), both indicating whether it is produced from natural gas or petcoke. It is expected that the fuel has a significant influence on the cost of production and the environmental profile of clinker production, but also on the sulfur balance, since petcoke is assumed to contain a 4% of sulfur. The content of ye'elimite was varied from 30% up to 70%, 10% ferrite and the rest balanced to be belite. As in previous analyses (T. Hanein et al., 2018b), a constrained optimisation of raw materials input was performed to minimise the input of bauxite. Sulfur to be fed to the kiln was assumed as pure elemental sulfur, produced in the Claus process for desulfurization of fossil fuels, and gypsum was assumed as pure calcium sulfate dihydrate. The amount of raw materials (clay, limestone, bauxite and gypsum) is provided in w/w of the raw meal, while the amount of sulfur is provided as mass of sulfur per total mass of clinker produced. The theoretical heat is the difference of the enthalpy of formation of

Table 2
Mass and energy balance of the studied CSA clinker formulations and reference cases.

Clinker case	Fuel	Content of ye'elimite	Source of sulfur	Clay	Limestone	Bauxite	Gypsum	Raw sulfur, kg per kg of clinker	Raw material/ Clinker, kg/t	Theoretical heat, MJ/t clinker	Assumed SO_2 raw emissions, ppm
				kg per kg of raw feed							
NG30	Natural Gas	30%	Sulfur	0.34	0.66	0	0	0.016	1466	982	1000
NG40		40%		0.29	0.65	0.06	0	0.021	1431	811	1100
NG50		50%		0.2	0.63	0.17	0	0.026	1395	670	1200
NG60		60%		0.11	0.61	0.28	0	0.031	1359	529	1300
NG70		70%		0.02	0.58	0.4	0	0.037	1323	388	1400
PK30	Petcoke	30%	Sulfur	0.34	0.66	0	0	0.012	1466	982	1100
PK40		40%		0.29	0.65	0.06	0	0.017	1431	811	1250
PK50		50%		0.2	0.63	0.17	0	0.023	1395	670	1400
PK60		60%		0.11	0.61	0.28	0	0.028	1359	529	1550
PK70		70%		0.02	0.58	0.4	0	0.033	1323	388	1700
REFCSA_NG	Natural Gas	60%	Gypsum	0.11	0.5	0.27	0.12	0	1429	1242	0
REFCSA_PK	Petcoke	60%	Gypsum	0.11	0.52	0.27	0.1	0	1431	1238	0
REFPC	Natural Gas	Portland	None	0.19	0.81	0	0	0	1666	1780	0
REFPC	Petcoke	Portland	None	0.19	0.81	0	0	0	1666	1780	0

products and the enthalpy of reactants, at 298 K, following a previous example (T. Hanein et al., 2018b). The assumed heat for the kiln process assumes 2000 MJ of losses per tonne of clinker, which is a standard value observed for Portland cement clinker when burnt at 1450 °C. This energy loss was also assumed for CSA making, although its clinkering temperature is significantly lower; the change in the consumption of fuels, exhaust gas flow and the different nature of the chemistry in the kiln has an unknown impact in the thermal efficiency. By assuming the same energy losses for CSA and for PC, a conservative approach towards the alternative product was introduced in the analysis.

All CSA production options using sulfur as a supplementary fuel were considered to produce a significant sulfur dioxide atmosphere. The emissions baseline was considered equal to a "high sulfur load" scenario of Portland cement as defined by the industrial data shown in the BAT reference document for the cement sector. In this case, the baseline for natural gas as fuel was set at 1000 ppm of raw SO_2 emissions for the lowest ye'elimite clinker, and up to 1400 ppm for the highest ye'elimite formulation; for petcoke, the range was varied from 1200 to 1700 ppm, since there is a higher load of sulfur to the kiln. Commercial CSA and Portland cement clinkers were assumed with no need of wet scrubbing in their manufacture. Further details on the mass and energy balance, along with the full set of results of the economic and environmental assessment, can be found in the Supplementary Material provided with this work.

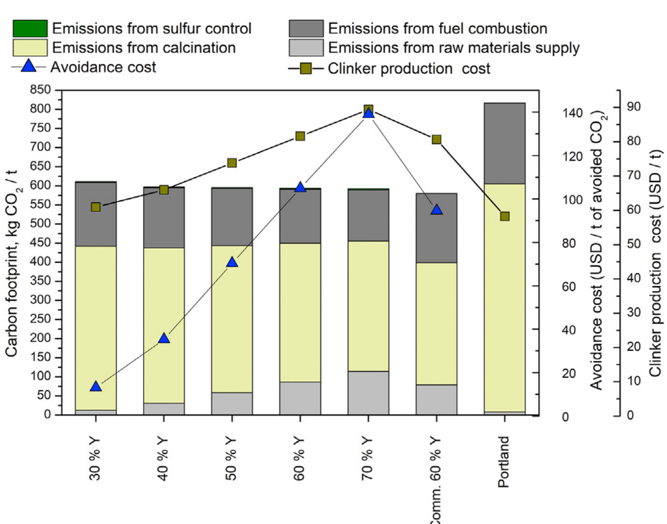

Fig. 2. Avoidance cost, associated emissions and clinker production costs as a function of the ye'elimite content, and compared to commercial CSA and Portland cement clinker production.

Fig. 2 shows the associated emissions of greenhouse gases emissions, measured as equivalent CO_2, the cost of clinker production and the total avoidance cost, calculated for life-cycle emissions as stated previously, for CSA with varying compositions made with sulfur, and commercial CSA and Portland cement clinker as reference cases. The fuel was assumed to be natural gas for all cases in this cart, and bauxite was assumed to be sourced from India. As observed, there is a large difference between all CSA options and Portland cement, since they all reduce the footprint from more than 800 kg CO_2eq per tonne of clinker down to values just above 600 kg CO_2eq. There is, therefore, a significant reduction of 25% of direct and indirect CO_2 emissions in the production of C.

Bauxite was assumed to be sourced from India, closer to Qatar than any other studied location. As per observation, the high impact of the transport of bauxite is compensated by the reduction of calcined limestone required in the formulation. Low ye'elimite cements have higher energy demand and clinkering temperature, and a higher amount of calcium carbonate in the initial meal, but the lower amount of bauxite compensates for the increased emissions regarding other formulations. When the ye'elimite content is higher, lower fuel is required, less amount of limestone needs to be calcined, but higher amounts of bauxite supply, which increases the emissions associated to its sourcing. This is an important feature of CSA cements, since indirect emissions, as those generated by the supply of raw materials, are significantly increased in comparison to conventional formulations. Remarkably, the total footprint of CSA cements in Fig. 2 seems constant at around 600 kg CO_2 per tonne of clinker, which is just an effect of the proximity of the source of bauxite. Closer distances would reduce the footprint associated to bauxite, i.e. the carbon footprint decreases as ye'elimite increases.

As expected, the control of sulfur with a wet scrubber has a small significance in the results from the operational point of view, but it is non-negligible. See the following sections for further analysis.

The cost of bauxite however is remarkably significant and influential in the economic balance of CSA cements. Only low ye'elimite CSA formulations would be competitive with PC at around USD 60 per tonne according to our estimations.

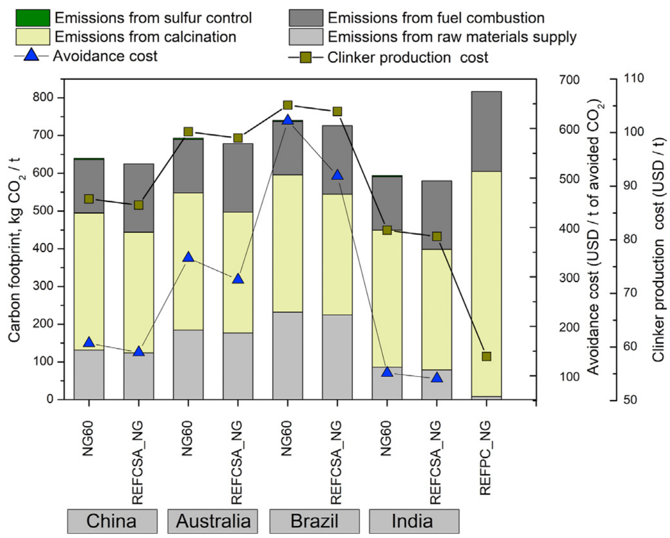

Fig. 3. Avoidance cost, associated emissions and clinker production costs as a function of the origin of bauxite. Formulations: NG60: natural gas, 60% ye'elimite, using commercial sulfur as supplementary fuel; REFCSA_NG: reference commercial CSA using natural gas as fuel; REFPC_NG: reference commercial Portland cement clinker using natural gas.

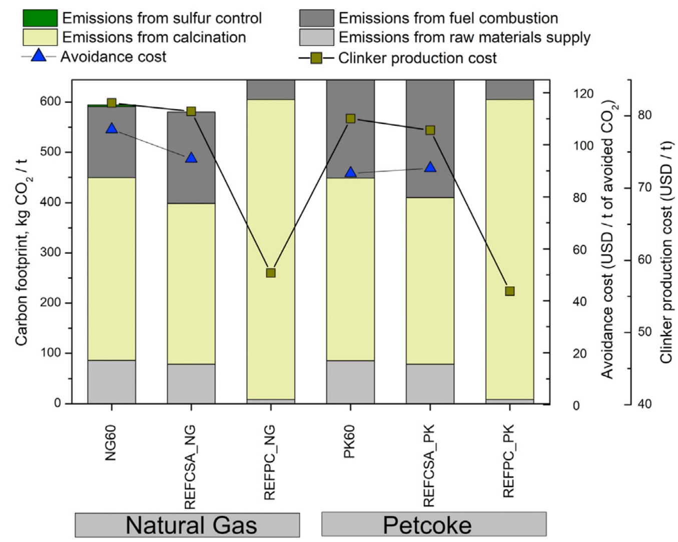

Fig. 4. Avoidance cost, associated emissions and clinker production costs as a function of the fuel used in the clinkering process. Formulations: NG60: natural gas, 60% ye'elimite, using commercial sulfur as supplementary fuel; REFCSA_NG: reference commercial CSA using natural gas as fuel; REFPC_NG: reference commercial Portland cement clinker using natural gas as fuel; PK60: petcoke, 60% ye'elimite, using commercial sulfur as supplementary fuel; REFCSA_PK: reference commercial CSA using petcoke as fuel; REFPC_PK: reference commercial Portland cement clinker using petcoke as fuel.

The CO_2 avoidance cost, which as stated before accounts for direct and indirect emissions, shows a rather low cost for low ye'elimite formulations when they substitute Portland cement clinker production. The minimum cost is observed at USD 13.1 per tonne of avoided CO_2 for CSA with 30% ye'elimite, and USD 35 for CSA at 40% ye'elimite.

3.2. Influence of bauxite origin, fuel and sulfur

Results show a similar sensitivity of the environmental and economic performance of CSA clinker production as observed in the literature (T. Hanein et al., 2018b). From Fig. 3 it can be deduced that, as expected, the distance from the source of Al_2O_3 source, bauxite, has an important influence on the environmental and economic performance of CSA clinkers with high content of ye'elimite. It is also observed that the performance of conventional CSA, with 60% of ye'elimite, has no significant difference with the process using sulfur burning for the same content of ye'elimite.

From Fig. 3 it can be observed that avoidance cost of CO_2, both for sulfur-burning or conventional CSA, is highly influenced by the origin of bauxite. All the options show a high avoidance cost, over USD 100 for bauxite from India and over 600 for bauxite from Brazil. The change of footprint from Portland is −27% for bauxite from India as maximum reduction, and - 9% for bauxite from Australia as minimum reduction (note that the label for each country refers to the locations shown in Table 1 for simplification). Costs of clinker production are importantly affected in cases with higher ye'elimite content. For example, in those cases shown in Fig. 3, commercial CSA would increase costs by 80% from PC clinker if bauxite is sourced from Australia, but 40% if sourced from India.

Therefore, there is an optimal distance range for supply of bauxite. However, from Fig. 4 it can be deduced that there is no optimum fuel in the evaluation of the avoidance cost, since the variations of greenhouse gases emissions are very similar for the two fuels. This provokes very similar avoidance costs of CO_2. On the other hand, the cost of CSA is slightly reduced when petcoke is used, but the associated emissions do anyway increase.

In Fig. 5, the methodological choice for sulfur is evaluated. In this chart, NG60W represents the formulation with 60% ye'elimite and natural gas as fuel, in which sulfur is considered burden-free from the environmental point of view, and it is compared to commercial CSA, Portland cement and NG60, in which sulfur is not free from environmental or economic burdens. It is noticeable the scarce effect of the methodological choice regarding the supply of sulfur. But, on the other hand, the avoidance cost per tonne of saved CO_2 is significantly affected by this methodological choice. In the case shown in Fig. 5, the variation of emissions from the base case to the alternative case are very small: lower than 1%. The cost variation is slightly higher, 4%. However, the combination of the two factors against Portland cement is highly significant, producing an

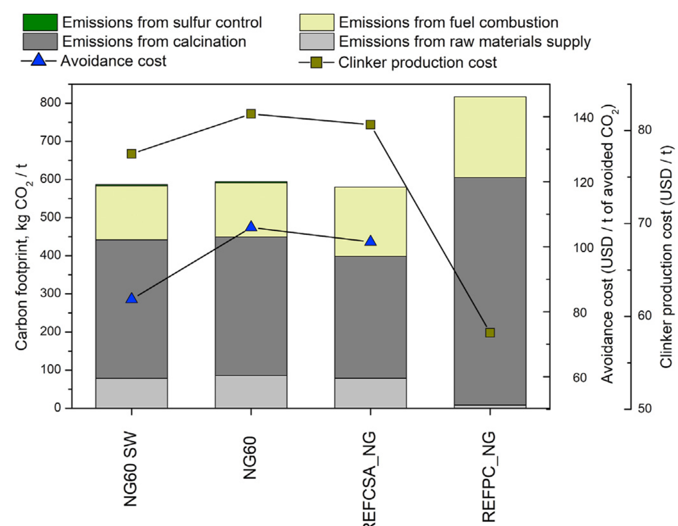

Fig. 5. Avoidance cost, associated emissions and clinker production costs as per methodological choices for sulfur. Formulations NG60 SW: natural gas, 60% ye'elimite, considering sulfur a waste; NG60: natural gas, 60% ye'elimite, using commercial sulfur as supplementary fuel; REFCSA_NG: reference commercial CSA using natural gas as fuel; REFPC_NG: reference commercial Portland cement clinker using natural gas.

Table 3
Direct CO_2 emissions from commercial CSA, and from CSA produced with sulfur burning.

Case	Portland	CSA	CSA
Cement type	Portland	Commercial CSA	CSA from sulfur burning
Sulfur Source	None	Gypsum	Sulfur
Ye'elimite content	None	40%	40%
Direct CO_2 kg/t clinker	808	565	567
Avoidance cost, USD/t of *direct* CO_2 emissions in the shift from Portland cement Clinker production	–	**28.2**	**20.1**

avoidance cost 21% lower when sulfur is considered a waste.

3.3. Wet scrubbing and prospective cost analysis

In our analysis, the CO_2 trading costs were not assumed in the economic balance, even though a significant amount of CO_2 emissions can be avoided if the production is shifted from Portland cement to CSA in the same kiln. In terms of emissions trading schemes or emissions taxing, the associated emissions to CSA cement manufacturing would go below emissions allowances and, of course, would have an impact on the economic balance through cost savings or tradable emissions. This would only affect direct emissions, which are of high relevance in the case of CSA cements. The raw materials mining and transport are excluded, so emissions trading or savings would have an important impact on the cost balance. An example of the direct CO_2 emission avoidance cost is shown in Table 3. In this case, we have used the formulation at 40% ye'elimite, which can be considered a competitive target formulation in terms of performance and CO_2 avoidance cost in the substitution of Portland cement.

As observed, both CSA formulations achieve a significant direct CO_2 reduction of around 30%. The shift of production avoids emissions at a cost over USD 28 per tonne of CO_2 for commercial CSA, while it is around USD 20 for the sulfur burning case. In this last option, costs may be higher if sulfur dioxide needs to be removed from exhaust gases through a wet scrubber, with a current operational cost of more than USD 2 per tonne of clinker. However, in the case of shifting a kiln towards CSA production with sulfur burning, it was assumed that, with no info on the performance of an industrially scaled sulfur burning cement kiln yet, the safest solution would probably be specified and a wet scrubber would be required as a precautionary measure towards pollution control.

In this section we propose a new prospective analysis of the evolution of costs of clinker production based on the need of a SO_2 scrubber and the evolution of CO_2 emissions taxing or trading towards 2050. In this economic projection the annual evolution of cost is calculated considering the following factors and assumptions:

- The avoided CO_2 IS monetised according to the potential savings in comparison to Portland cement production to the year 2050. In this way, the accountable emissions for trading or savings are calculated from the Portland cement baseline, which is assumed to evolve according to the 2DS (2° scenario) scenario of the cement industry roadmap, i.e. to a 24% reduction in 2050 (Cement Sustainability Initiative and International Energy Agency, 2019). The chargeable tax or price of CO_2 emissions will be assumed equal to that assumed by van Ruijven et al. (2016), in which a current moderate climate policy scenario in 2020 evolves towards a cost of USD 65 per tonne of emitted CO_2 in 2050. All the prospective dynamic models are assumed linear, i.e. costs evolve linearly with time.
- The required investment of a wet scrubber in the cement kiln shift for CSA clinker production would consist of USD 6 million in an installation producing 1000 tonnes of clinker per day. An annualization of the investment cost for three years was assumed, i.e. the cost of clinker pays back for three years to compensate for the capital expenses in the initial year.
- Inflation of raw materials was assumed at 1.5% per annum, while 0% was assumed for energy sources due to the existence of high uncertainties. This assumption reduces the potential savings from CSA options, with a lower use of fuel, and it constitutes a conservative approach in the estimation of costs.

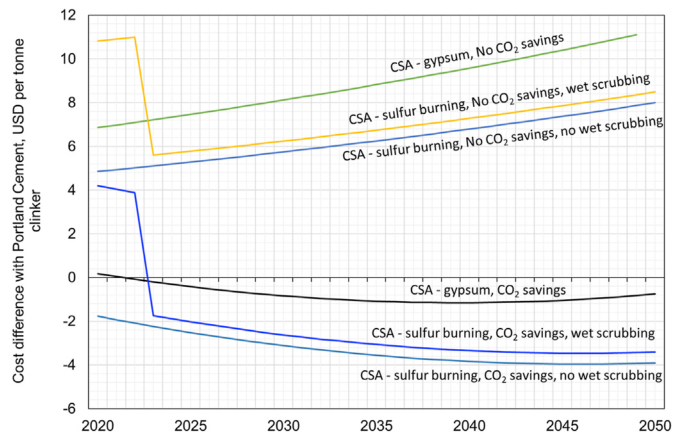

Fig. 6. Prospective cost difference with Portland Cement of CSA options using sulfur burning or conventional (gypsum) as source of sulfur, with and without wet scrubbing, and with and without CO_2 savings considered in the cost balance.

In total, six curves can be drawn for the evolution of costs, represented as the cost difference with Portland cement clinker in Fig. 6:

- Two curves for commercial or conventional CSA, one with CO_2 savings being considered and one without considering them.
- Four curves for sulfur-burning- CSA, in which the four combinations of CO_2 emissions savings and the need for wet scrubbing are incorporated to the economic balance.

As observed, time is a relatively detrimental factor for CSA production when CO_2 related savings are not taken into account. The assumed annual increase of raw materials costs has an important impact in the more expensive raw materials of CSA production. Wet scrubbing increases the cost by *ca* USD 5 per tonne of clinker (around 5–10% of total clinker cost production) during the payback time, introducing a period of higher unit costs in the initial stages of CSA implementation.

The influence of costs savings generated from the reduction of emissions of greenhouse gases is remarkably important. In the case of commercial CSA, the reduction of costs is at USD 7 per tonne of clinker, increased to USD 12 in 2050. In the case of CSA made with sulfur burning, the influence of CO_2 – related cost savings is essential to achieve competitive production costs, as observed in Fig. 6. Therefore, in an optimistic scenario, with no need for wet scrubbing, and with a consideration towards CO_2 savings in the form of environmental cost credits, the economic balance of the system would favour the production of CSA clinker with low ye'elimite using sulfur as a fuel and a raw material.

4. Discussion

Our study focuses on a new technology for CSA production method consisting on the combustion of sulfur to supply SO_3/SO_2 to the reaction. This new approach is proven to reduce the associated greenhouse gases emissions of Portland cement clinker production. However, CSA clinker production costs are higher than those for Portland cement clinker, even for low ye'elimite compositions. The closest locations of bauxite sources might ease the economics of production. In this context, the shift of traditional Portland cement kilns to low ye'elimite CSA clinker production has been proven as an effective method for the reduction of GHG emissions associated to Portland cement clinker production. And, despite the generally observed increased costs of clinker production, the shift to low ye'elimite CSA cements has the lowest costs per tonne of avoided CO_2 in the industry, much lower than the currently commercial CSA or technologies for carbon capture, such as post-combustion capture with amines or oxycombustion of fuel.

Without the aim of undermining the scope of this work, these conclusions were probably well known by the cement industry, and that is proven through the effort that the cement industry is making in the development of alternative formulations of cement with low associated emissions (Nidheesh and Kumar, 2019); this paper can help to understand the interaction between most of the techno-economic variables around CSA clinker production and how it can actually be a location-driven solution in certain markets.

There are, however, certain aspects around the analysis above that have a strong influence on the results and on the associated uncertainty: (i) the comparability of the performance of CSA cements to that of Portland cements; (ii) sourcing of bauxite, which poses questions on the geographical limitations in the supply of bauxite, and on the proven reserves to sustain an economically viable CSA production scheme over the years; (iii) trading-off of environmental impacts, since a higher sulfate-bearing environment within the kiln could potentially lead to higher SO_2 emissions.

4.1. On the performance of CSA cements

The most evident aspect for discussion is the question on the clinker functionality assumed in this work, as 1 tonne of Portland cement clinker is compared with 1 tonne of CSA clinker. First, it is better to emphasise the understanding of functionality used in this paper: we are benchmarking cement clinker production in a cradle-to-gate approach, looking at the emissions associated to a production system, i.e., the function is *to produce clinker*. However, the further manufacture of cement, concrete and how different chemistries of clinker affect the performance of cementitious products are not considered in our scope. Although the performance of final products is not compared, our data can easily feed the comparison of cement-based materials to be made by other analysts, where the performance of new products can be compared to reference Portland cement materials, such as concrete, mortars and other applications (Murray et al., 2019; Pera and Ambroise, 2004). In this sense, *what is known* about Portland cement has an important advantage if compared to *what is known* about CSA cements: Portland cement is well-known after more than 100 years of experience and research, which has focus on its sustainability for the last 50 years, while CSA has not been considered a potential massive substitute of Portland cement until recent times (Gartner, 2004; Gartner and Sui, 2018), and its standardisation and development towards the market is still pending in the case of low ye'elimite compositions. This fact leaves, at least, two open questions: (i) can low ye'elimite CSA cement be considered a substitute for all, most or some of the applications of Portland cement? (ii) Do the current improvement options for Portland cement (e.g. use of substitute materials, alternative fuels, mix with alternative binders, etc.) have an equivalent impact in CSA cements?

4.2. On the geographical limitations

In a previous work, we discussed the suitability of the location of the manufacturing site for the eco-efficient production of CSA, within the framework of standardised definition of eco-efficiency (Gálvez-Martos et al., 2020; ISO, 2012), where a product value is related to its environmental performance in a life cycle perspective. It was found that CSA cements can be considered eco-efficient only in certain circumstances; the distance to the source of bauxite is key to enable the eco-efficient character of CSA clinker. In addition, in this study, the concept of avoidance cost sets a benchmark for the shift of kilns and concluded that only low ye'elimite is suitable for mass production of ordinary Portland cement substitutes. The results show that the preferred CSA option should focus on low ye'elimite, not only due to the high environmental and economic impact derived from the supply of bauxite (extraction and transport), but also the increased pressure over proven reserves in places such as Australia, Brazil, India and others (USGS, 2018). Although world aluminium demand is increasing, the proven reserves would suffice to satisfy the market (USGS, 2018); also, the industry is extensively recycling aluminium to alleviate the economics of its production. The effect of the massive production of CSA cement using bauxite as raw material is uncertain on bauxite price and market. As a proxy indicator, if world cement demand, currently at 4.2 billion in 2019, is shifted to 40% ye'elimite formulation, it would require doubling the production of mined bauxite, i.e. from the $3 \cdot 10^9$ tonnes extracted currently (USGS, 2018), to more than $6 \cdot 10^9$. Regarding our specific analysis, we have observed that in the specific situation of Qatar a rather environmentally optimal situation takes place for CSA production, where the foreseen healthy demand of cement in the next 10–20 years makes cement also an optimal showcase for alternative formulations.

4.3. On environmental trade-offs

Qatar and the Middle East make a perfect showcase for the third question in this discussion: the use of sulfur as a fuel and as a precursor of sulfate for the final formulation. In Qatar, sulfur from desulfurization operations is readily available, which upon oxidation would provide the sulfate required in the final formulation of CSA cement. Sulfur barely reaches 3.7% in formulations with high ye'elimite. But, even in that case, the amount of sulfur (or sulfate) to be fed to the kiln is several times higher than the amount of sulfur (normally as an impurity) entering into a Portland cement clinker kiln through the fuel (e.g. petcoke). As stated before, the alkaline nature of the kiln makes the process to easily absorb sulfur dioxide and trioxide emissions; if the excess is fed through calcium sulfate, the stoichiometry of the process ensures its absorption due to the compensation of extra oxygen and CaO. Feeding sulfur in the reduced elemental form needs an extra supply of oxygen for the formation of the required sulfates, and the chemistry of the kiln should favour absorption through an alkaline environment. In our analysis, which has followed a conservative approach, the results considered a wet scrubber in order to calculate a wider spectrum of emissions and operational costs; and an increased production cost of around 2–3% was calculated when raw emissions are in the range of 1000 to 1700 ppm of SO_2. Whether these assumptions were oversized or not, depends on the actual technology of the kiln, the number of preheaters, and the quality of solid-gas contact. The Best Available Techniques document for the cement sector in Europe reported raw emissions at a maximum of 1700 ppm of SO_2 before scrubbing for high sulfur environments, i.e. with raw meal content of 2–4% sulfur, which means that a large proportion of the sulfur input is captured and absorbed as sulfate (European Commission, 2013), while pilot trials using sulfur burning have detected larger raw emissions of SO_2, but in pilot kilns without preheaters, precalciner and with a poor mass transfer (Hanein et al., 2016).

The short-term economics of CSA production depend on the need of controlling emissions of pollutants. In fact, a relatively short increase of production costs by 8–10%, during the payback time, would be required to achieve an economically neutral operation of CSA production. This fact could negatively drive the short-term investment in such technology. But a long-term strategy that seeks to monetize the achievable greenhouse gases savings, e.g. through tradable CO_2 savings, would compensate the shift of production towards novel formulations with a better environmental profile but similar production costs. The expectable more ambitious policies and schemes derived from the Paris Agreement may determine a faster deployment of alternative formulations such as CSA cements in a much faster manner than what can be expected from a purely static cost analysis.

5. Conclusions

The techno-economic performance of CSA clinker production was analysed and proven as a cost-efficient opportunity for a low carbon cement industry, provided that certain conditions are met. First, due to the nature of the formulation of CSA, the content of ye'elimite determines the amount of Al_2O_3 to be fed into the process as bauxite, which introduces higher costs and environmental impacts from its supply. Second, the fuel has an important influence in the total amount of CO_2 from clinker production but has no influence on the avoidance derived from the shift from conventional Portland cement to CSA. Third, the use of sulfur burning has certain benefits associated with lower CO_2 avoidance cost but may imply higher costs derived from SO_2 emissions control through wet scrubbing.

In this assessment, the geographical scope included the specific circumstances of Qatar, which strategic position for bauxite imports, the availability of sulfur, and the high cement demand would make this country a perfect demonstration case for a low ye'elimite CSA massive production site. In the light of the new Paris Agreement, the framework of a more demanding environmental policy towards carbon emissions and climate change mitigation may help to the further development of low carbon cement formulations such as CSA all over the world.

CRediT authorship contribution statement

José-Luis Gálvez-Martos: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Roneta Chaliulina:** Methodology, Formal analysis, Investigation, Writing – review & editing. **Ammar Elhoweris:** Conceptualization, Investigation, Writing – review & editing. **Jonathan Mwanda:** Supervision, Writing – review & editing, Formal analysis. **Amer Hakki:** Supervision, Resources, Writing – review & editing. **Yousef Al-horr:** Supervision, Conceptualization, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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