



Integration of oxygen membranes for oxygen production in cement plants



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ABSTRACT

The present paper describes the integration of oxygen membranes in cement plants both from an energy, exergy and economic point of view. Different configurations for oxygen enrichment of the tertiary air for combustion in the pre-calciner and full oxy-fuel combustion in both pre-calciner and kiln are examined. The economic figures of merit are compared with those from a standard cryogenic plant. Both oxygen enriched air and full oxy-fuel cases allow for an increase in clinker production, use of alternative fuels as well as on-site electricity production. In addition, the full oxy-fuel cases generate a concentrated CO₂ source that can be used for enhanced oil recovery, in combination with biomass gasification and electrolysis for synthesis gas production, or possibly sequestered. The cases with oxygen enriched air provide very promising economic figures of merit with discounted payback periods slightly higher than one year. The full oxy-fuel cases have a discounted payback period of approximately 2.3 years assuming a CO₂ selling price of 35 US\$/ton. The sensitivity analysis of full oxy-fuel cases clearly shows that for the discounted payback period, the most sensitive parameters are the CO₂ price and the clinker selling price.

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1. Introduction

The cement industry is one of the main carbon dioxide emitting industries in the world, constituting approximately 5–7% of the global anthropogenic carbon dioxide emission [1]. The process emits, per ton of cement produced, around 900 kg CO₂ [2], of which approximately 50% come from the conversion of limestone (CaCO₃) into CaO (calcium oxide); another 40% are due to the fuel combustion in the kiln, 5% from transportation and the remaining 5% is from the electricity consumption [3,4]. Modern cement plants have drastically reduced the fuel consumption by heat integration of all subsystems, and in many cases, replaced fossil fuels by biomass or waste [5,6]. Thus the only option to significantly reduce greenhouse gas emissions in the cement sector is CCS (CO₂ capture and sequestration) [7]. Different studies [3,7] have pointed out that fossil fueled cement production is a very important process to

consider in CO₂ emission reduction plans because each mole of pure O₂ generates 2–3 times the amount of CO₂ generated in power production, due to the calcination of limestone [8].

Post-combustion capture and oxy-fuel combustion technologies have been proposed as possibilities for CO₂ separation in cement plants [9–11]. Pre-combustion is not suitable because it is unable to capture the emissions from the conversion of limestone to lime. Post-combustion capture technology is considered a suitable short-term option for CCS in the cement sector because it doesn't require fundamental changes in the clinker-burning process, and could be retrofitted to existing plants. It relies on the downstream separation of CO₂ using different chemical or physical measures like chemical absorption (amine scrubbing, chilled ammonia), membrane technologies, adsorption technologies, mineralization and calcium looping [12]. The most mature and proven post-combustion technology to separate the CO₂ is chemical absorption using amine-based solvents. The CO₂-rich solvent is then regenerated in a stripping process by addition of low-temperature heat, typically low-pressure steam. The energy requirements for the regeneration of the amines are high and additional installations such as a cogeneration plant are usually needed. In addition to the

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heat requirements for the solvent process, other challenges for post-combustion technologies are related to managing the dust and SO_x emissions. Other post-combustion capture techniques, like membranes and solid sorption processes are in a research state and may lead to less energy-intensive capture options. A relatively new and upcoming carbon capture process is the carbonate looping process [13–16], also known as the regenerative carbonate cycle. Unlike the amine and ammonia processes, a dry limestone-based absorbent is used to capture CO_2 . This technology uses DFB (dual fluidized bed) reactors (i.e. the carbonator and the calciner) with a continuous looping of Ca-bearing solids. The capture principle is based on the reaction of lime (CaO) with CO_2 , forming CaCO_3 (calcium carbonate), at a temperature range between 600 °C and 700 °C, in the carbonator and the reverse reaction in the calciner (900 °C), producing a rich- CO_2 gas stream and a quantity of calcined lime, which is active for further CO_2 absorption. The calciner has to be operated under oxy-fuel conditions to prevent the dilution of the captured CO_2 with nitrogen; the oxygen demand is about 40% in relation to the oxygen demand of an oxy-fuel kiln [17].

Oxy-fuel technology seems to be the most promising of the CCS technologies in the long term perspective [10] because of the low oxygen consumption (compared with power plants) and the high concentration of CO_2 due to calcination. It can give efficiency gains in cement production and improve the energy demand but still needs further research and requires changes in the core cement production process that make it more suitable for new cement plants.

In full oxy-fuel combustion, nitrogen is eliminated and the resulting flue gas stream is highly concentrated in carbon dioxide, which can be compressed and liquefied for underground sequestration or used in EOR (enhanced oil recovery) applications [18]. Even if the valorization of CO_2 in EOR is highly debatable due to elevated investment costs, CO_2 transportation, energy consumption, plant operability and environmental and long-term consequences; it should also be noticed that in a future energy system, dominated by renewables, concentrated CO_2 sources can actually become valuable and of high interest. CO_2 can be used in combination with biomass gasification and electrolysis of steam to produce a synthesis gas that can be upgraded to liquid fuels.

Oxygen enrichment of air is less potent, compared with full oxy-fuel combustion, but an economically interesting method for reducing the carbon footprint of cement production. The method is well known in the industry to increase the production capacity [9–11] and the use of alternative fuels [12]. Many cement plants are limited by their induced draft fans that reach their limits before other equipment or systems tied to the kiln. However, increasing the production requires additional fuel and air for the combustion. In these cases, oxygen enrichment provides the necessary additional oxygen for combustion without the volume penalty of air. In addition to the immediate higher clinker production, oxygen promotes a more stable operation by creating a hotter, shorter flame and improving burning zone control. Over time, this added stability contributes to additional production. Some studies [19,20] have quantified the benefits of oxygen enrichment of air in the kiln burner. An increase of 25%–50% of kiln capacity (short term experiments) was reported for an air oxygen enrichment of 30–35 vol.% [19]. The same report indicates that oxygen enrichment can reduce fuel consumption between 100 and 200 $\text{MJ}/t_{\text{clinker}}$ but, at the same time, the electricity consumption increases between 10 and 35 $\text{kWh}/t_{\text{clinker}}$ due to oxygen production. The modeling results of Duan et al. [20] estimate that when the O_2 concentration is increased from 21% to 30%, the coal consumption decreases by 18%, the flue gas volume reduces by 13% and the CO_2 concentration in the flue gas increases to 77%. Regarding air oxygen enrichment in the pre-calciner, potential capacity improvements of

3.5 and 4 $t_{\text{clinker}}/t_{\text{extraO}_2}$ have been reported in Refs. [21] and [22], respectively.

The costs of oxygen production are a significant barrier to the application of full oxy-fuel combustion and oxygen enrichment. Large scale oxygen separation from air is generally achieved by cryogenic distillation, which is a mature technology that allows for high purities (>99%). However, it is a very energy intensive process operating at very low temperatures and elevated pressures. The energy input to generate one ton of oxygen (>99%) has been reported to be, for large scale production plants, 220 $\text{kWh}/t_{\text{O}_2}$ [23], 240 $\text{kWh}/t_{\text{O}_2}$ [24] and 245 $\text{kWh}/t_{\text{O}_2}$ [25]. A promising alternative to cryogenic distillation is MIEC (mixed ionic and electronic conducting) oxygen membranes (sometimes referred to as OTMs), operating at high temperatures (usually >700 °C). The driving force for the separation is an oxygen pressure difference across the membrane. High purity oxygen can be obtained because only oxide ions can be transported through the membrane. The working principle of the membrane is as follows: oxygen is adsorbed on the feed side, subsequently split and ionized and then diffuses through the membrane in ionic form (Fig. 1). To preserve electro-neutrality, there is a simultaneous flux of electrons in the opposite direction. The oxygen can be extracted as a pure product and nitrogen, in principle, would only enter via pinholes or defects in the sealing. The energy consumption for oxygen production using MIEC (mixed ionic-electronic conducting) membranes is anticipated to only depend weakly on the plant size, and values of 185 $\text{kWh}/t_{\text{O}_2}$ [26] and 147 $\text{kWh}/t_{\text{O}_2}$ [27] have been reported from system/BOP simulation studies.

A significant benefit of ceramic membrane technology is that it allows for thermal integration in the process, so the energy consumption numbers can become significantly lower [28,29]. For instance, when supplying oxygen for a biomass gasifier in a thermally integrated arrangement the power need may be as low as 100 $\text{kWh}/t_{\text{O}_2}$ [28]. Future prospects reported for the membrane route to pure oxygen include [30,31]:

- Potential for 25–35% reduction in capital requirements over conventional cryogenic plants.
- A 30% reduction in operating costs for the process.
- 35–60% reduction in power consumption (depending on product pressure) up to 1000 psig.
- No net electricity consumption in certain applications.

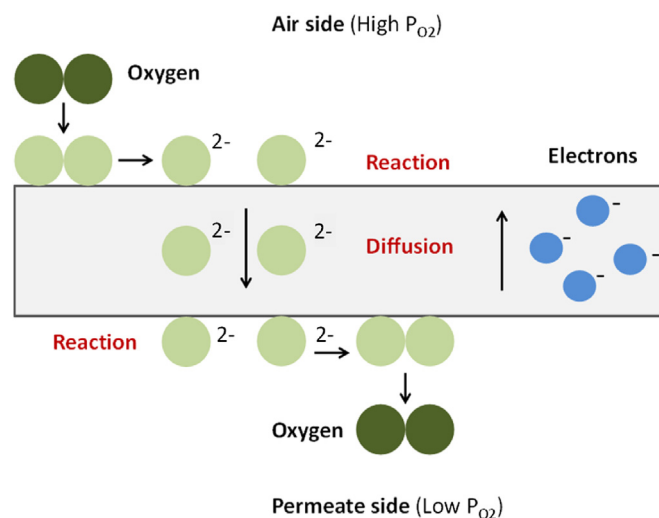


Fig. 1. Schematic of the oxygen transport mechanism in a mixed ionic-electronic conducting (MIEC) membrane.

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