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Utilization of blast furnace flue gas: Opportunities and challenges for polymeric membrane gas separation processes



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ABSTRACT

New programs seeking to improve energy efficiency and reduce GHG emissions from the steelmaking industry are studying alternative uses for blast furnace gas such as the VALORCO project in France. One promising alternative is CO_2 and CO utilization in which emissions are used for the production of valuable products, like industrial intermediates and fuels synthetized from CO or CO_2 or both. A gas separation and concentration technology is necessary to recover industry emissions and produce gas stream(s) suitable for carbon utilization technologies. This paper describes the application of commercially available polymeric gas separation membranes to blast furnace gas. Data for two commercially available membranes, a H_2 selective glassy polyimide membrane from UBE industries and a CO_2 selective rubbery composite membrane from MTR Inc. along with a process economic modelling have been used for the calculation of product composition paths and associated costs depending on product purity and recovery. Relatively low separation costs, from 15 to 33 $\mathrm{EUR}/$ Ton separated CO_2 , seem promising for the use of membrane technology as a concentration tuning box prior to carbon utilization technologies, notably for mid to high CO_2 purity requirements, and allow to simultaneously achieve CO_2 capture and CO recovery.

1. Introduction

Industry is responsible for approximatively one-third of global final energy use and around 40% of total energy-related CO₂ emissions. Despite continuous efficiency increase in energy utilization, overall growing industrial production has led the CO₂ emissions to rise [1]. This is particularly true for the iron and steel industry, where the production increase in the past two decades is closely related to the development of emergent economies such as China and India. Global steel production almost doubled between 2000 and 2014, going from 0.85 MTons/year to 1.67 MTons/year [2]. GHG emissions depend on the production technology. Two production technologies account for most of the steel produced globally: Integrated steel mills based on the Blast Furnace-Blast Oxygen Furnace (BF-BOF), and the Electric Arc Furnace based on recycled steel or scrap. Integrated steel mills dominate global production with over 70% of crude steel worldwide being produced by this process and the increase in steel production over the past decades was mainly achieved by an increase of production by this technology [2]. BF-BOF technology consumes between 13 and

14 GJ/ton of steel and can produce up to 1.9 t of CO₂ per ton of steel depending on energy efficiency and the percentage of carbonated energy according to local conditions [3].

Gas emissions in a conventional steel mill come mainly from three streams: Coke oven gas, blast furnace gas and blast oxygen furnace gas. Fig. 1 shows a simplified flowsheet of a conventional mill illustrating these three sources.

Blast furnace gas (BFG) emissions are the most important in volume and represent around 69% of the emitted CO_2 . BFG is mainly composed of 20-28% CO, 17-25% CO_2 , 50-55% N_2 and 1-5% H_2 [3]. It is typically burned for heating and power generation often being mixed with natural gas, coke oven gas or blast oxygen furnace gas to increase its heating value.

Energy and emissions concerns have led to research programs seeking to improve energy efficiency and reduce GHG emissions from the steelmaking industry. Recent works have studied the sequestration of $\rm CO_2$ from iron and steel off gases [4–9]. New programs are studying alternative uses for blast furnace gas in order to reduce emissions such as the VALORCO project in France. One promising alternative is $\rm CO_2$

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Nomenclature		MF_{HX}	Heat exchanger material factor [Dimensionless] Thickness of the dense membrane layer carrying the
а	Annuity coefficient for equipment [Dimensionless]	ι	separation [m]
a_m	Annuity coefficient for membrane module	OPEX	Operational expenditures [EUR/year]
m	[Dimensionless]	P_{cpr}	Compression power consumption [kW]
A_H	Heat exchange surface [m ²]	$\stackrel{r}{PF}$	Pressure factor [Dimensionless]
A_m	Membrane area [m ²]	P_{i}	Permeability of component i in the membrane [mol.m/
CAPEX	Capital expenditures [EUR]	ı	(m2.s. Pa)]
C_{cap}	Annual capital costs [EUR/year]	$\overline{P_i}$	Permeance of component i [mol/(m2.s. Pa)]
C_{CO2}	Specific CO ₂ separation cost [EUR/Ton CO ₂]	P_{mf}	Membrane frame pressure [bar]
C_{cw}	Annual cooling water cost [EUR/year]	P_{out}	Outlet gas pressure [bar]
C_{en}	Annual electricity cost [EUR/year]	P_{tot}	Total energy consumption [kW]
C_o	Compressor base cost [USD]		Vacuum power consumption [kW]
$C_{\text{O&M}}$	Annual operation and maintenance investment cost	$\frac{P_{vp}}{\overline{P_0}}$	Permeance of a base component (fastest) [mol/(m2.s. Pa)]
occ.n	[EUR/year].	p'	Membrane module upstream pressure [Pa]
C_{tot}	Total annual costs [EUR/year]	p''	Membrane module downstream pressure [Pa]
C_{vp}	Vacuum pump cost factor [EUR/kW]	$P_{i(0)}$	Partial pressure of component i at the feed side of the
D_i^r	Diffusion coefficient of component i in the polymer	- 1(0)	membrane [Pa]
	membrane [m2/s]	$P_{i(l)}$	Partial pressure of component i at the permeate side of the
ER	Exchange rate [EUR/USD]	1(1)	membrane [Pa]
HX_o	Heat exchanger base cost [USD]	Q	Gas flowrate along the upstream side [mol/s]
ICF	Indirect cost factor [Dimensionless]	Q_f	Feed gas flowrate [mol/s]
I_{Hex}	Heat exchanger investment cost [EUR]	$\vec{Q_p}$	Permeate gas flowrate [mol/s]
I_c	Compressor investment cost [EUR]	q	Dimensionless flow [Dimensionless]
I_m	Membrane surface investment cost [EUR]	S	Dimensionless membrane surface [Dimensionless]
I_{mf}	Membrane permanent frame investment cost [EUR]	t_{op}	Operation time per year [h/year]
I_{vp}	Vacuum pump investment cost [EUR]	UF	Update factor [Dimensionless]
J_{i}	Molar flux of component i permeating trough the mem-	W_{tot}	Total water consumption [m ³ /h]
	brane [mol/(m2s)]	X	Molar fraction at the upstream side [Dimensionless]
K_{cw}	Cooling water cost factor [EUR/m³]	x_f	Feed gas mole fraction [Dimensionless]
K_{el}	Electricity cost factor [EUR/kWh]	x_o	Retentate gas mole fraction [Dimensionless]
K_i	Sorption coefficient of component i in the membrane	У	Molar fraction at the downstream side [Dimensionless]
***	[mol/(m3Pa)]	y_p	Permeate gas mole fraction [Dimensionless]
K_m	Unit cost of membrane module [EUR/m ²]	α_i	Ideal membrane selectivity [Dimensionless]
K_{mf}	Base frame cost [EUR]	θ	Dimensionless stage cut of the membrane separation
K_{mr}	Membrane replacement cost [EUR/m²]		[Dimensionless]
$M_{CO2year} \ MDF_C$	Annual separated CO ₂ [Tons/year] Compressor module factor [Dimensionless]	ν	Membrane annual replacement rate [Dimensionless]
-	Heat exchanger module factor [Dimensionless]	Ψ	Pressure ratio [Dimensionless]
MDF_{HX} MF_C	Compressor material factor [Dimensionless]		
IVII'C	Compressor material factor [Difficustomess]		

and CO utilization in which emissions are used for the production of valuable products, like industrial intermediates and fuels synthetized from CO and or CO₂, either by chemical or biological transformations. This alternative would need a low energy, clean, safe separation technology (or technologies) capable of treating high volumes at economical rates.

Technologies typically applied for CO₂ and CO separation share common characteristics in their operation, the main difference being the separation agent itself and the nature of its interactions with the gas species to be separated. These separation processes include chemical absorption by amine systems in the case of CO₂ retention, and by copper salt solutions for CO; physical absorption for CO₂ by selexol or Rectisol processes; adsorption processes like pressure swing adsorption (PSA) or vacuum pressure swing adsorption (VPSA) over CO₂ and/or CO selective adsorbents; cryogenic separation, and membrane separation. To our knowledge, only two recent research programs have proposed separation technologies for the specific case of the iron and steel industry and have carried out tests on pilot level installations, both within CO₂ capture and storage scenarios. The european ULCOS

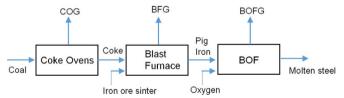


Fig. 1. Simplified flowsheet of an integrated steel mill.

program proposed a modification of the Blast Furnace, by which the blast furnace is fed by an oxygen stream instead of air thus eliminating the N_2 in the off-gas. This off-gas goes to a capture unit for CO_2 and the residual gases, containing CO, are recycled back to the BF as reducing gases. This is known as a Top Gas Recycled Blast Furnace (TGR-BF). For the capture step, a combination of PSA and cryogenics in order to achieve the purity necessary for CO_2 storage was proposed [8]. The Japanese COURSE 50 program has proposed a PSA process in which

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