

POST-COMBUSTION CO₂ CAPTURE: A COMPARISON BETWEEN COMMERCIALLY READY TECHNOLOGIES

Stefano E. Zanco, Jose-Francisco Pérez-Calvo, Viola Becattini and Marco Mazzotti*

Institute of Process Engineering, ETH Zurich, Sonneggstrasse 3, Zurich, 8092, Switzerland

* Corresponding author e-mail: marco.mazzotti@ipe.mavt.ethz.ch

Abstract

Retro-fitting of post-combustion CO₂ capture units to coal fired power plants is key to the transition to a net-zero CO₂ emission reality. Different separation technologies have been found suitable for CO₂ capture, but a more comprehensive approach is required to identify the most viable option among those commercially available. In this study we analyze the three most established technologies, namely absorption, adsorption and membrane separation, comparing both their exergetic efficiency and their total cost. This assessment provides an overview of the technical differences among the three capture routes and a realistic estimate of the expenditures associated with post-combustion CO₂ capture, as of today.

Keywords: Post-combustion CO₂ capture; techno-economic analysis; piperazine; TSA; membranes

1. Introduction

Pursuing the global goal of zero carbon-emissions by 2050 and keeping global warming below 1.5°C, carbon capture technologies offer a way to decarbonize a range of sectors, including the energy and the heavy industry sectors, where it is proving difficult to meaningfully reduce emissions in a near future[1,2,3].

More specifically, CO₂ capture via a post-combustion capture system represents the most immediate and viable option to retrofit existing plants, as it represents the only possibility to reduce the emissions at the stack without affecting the manufacturing process[3,4]. As regards post-combustion systems, the availability of a range of commercial-ready technologies suitable for point sources of different size and typology is crucial to reduce the price of the emission reduction, thus speeding up the

transition toward a net-zero-emission reality, before the complete decommission of fossil fuel combustors.

Amine wash is acknowledged as the least energy intensive among post-combustion technologies[3], and it is the only one whose operability has been already proven through pilot testing on a variety of scales[5,6]. Adsorption-based and membrane-based processes have been instead broadly studied and tested at lab or pilot scale. They benefit from a more basic process structure, where fewer sub-processes and devices are involved[7]. Although each one of these components is in general expensive (both in terms of capital and operational costs) and their cost tends to scale up linearly with the plant size, the price of the avoided CO₂ can be positively affected by the overall process simplicity. However, both technologies suffer from a strong limitation: Both commercial sorbents and commercial membranes for CO₂ separation cannot cope with the moisture of the flue gas, thus requiring a pretreatment unit for dehydration (Figure 1).

With this contribution, we aim at ranking the performance, in terms of both efficiency and capture costs, of adsorption and membranes in comparison to the most established absorption technology. The twofold nature of the detailed techno-economic analysis we have performed allows us both to highlight the differences in the final results, and to justify them consistently with the intrinsic features of the chemical separation processes involved.

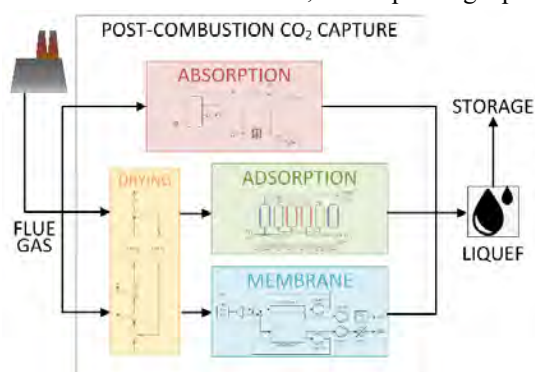


Figure 1. Schematic representation of the three post-combustion capture alternatives investigated within a general CCS system. The coal combustion flue gas is conveyed to the carbon capture plant, after a dehydration treatment in the case of adsorption or membrane. After the separation, CO₂ is ready to be liquefied and then transported to storage.

2. Methods

To compare the three technologies, an example case of post-combustion application has been chosen. Our reference emitter consists in a large point source, which

releases a flow rate of 10'000 tons per day of flue gas with a concentration of 12% vol. CO₂ (dry based), typical of a coal fired combustion. The capture units considered are required to meet those separation performance that are commonly assumed for Carbon Capture and Storage (CCS) applications, i.e. a rate of CO₂ recovery of 90% and a final CO₂-product purity of at least 96%.

A very straightforward path to the identification of the most suitable process design for each technology would be that of optimizing the process design subject to the minimization of the total plant cost. This approach has though a major drawback. In fact, the universality of the assumptions required to perform a cost analysis is rather limited, and the uncertainties deriving from the specificity of each assumption might impact the definition of an optimal design that could be considered as such from a more general prospective.

For this reason, in this study a two-step approach is preferred:

- First of all, each separation process is optimized from a merely technical point of view, aiming at maximum plant productivity and minimum exergy consumption, which can be reasonably considered technical proxies of the Capital Expenditures (CapEx) and Operational Expenditures (OpEx), respectively, associated with the realization and operation of a capture plant. This two-objective optimization yields a set of equivalent optima (a Pareto set).

- In a second step, the process designs forming the Pareto set are screened by means of a detailed cost analysis, assessing the CapEx and OpEx involved, estimating the total cost of capture, in terms of monetary annualized cost per ton of avoided CO₂ emission, and thus identifying the single economic optimum among the Pareto set.

Detailed rate-based models have been deployed for the simulation of the core process of each technology (i.e. the absorption columns, the adsorption column and the fibers of the membrane modules), and the cost estimations have been extended to include all the equipment involved in the CO₂ capture process[8,9,10,11].

Table 1. Boundary conditions for the operation of the CO₂ capture unit.

Parameter	Unit	Value
Flue gas composition	%	12/88 CO ₂ /N ₂
Flue gas relative humidity	%	95 @ 30°C
Flue gas temperature	°C	30
Flue gas pressure	bar(a)	1.3
CO ₂ recovery rate	%	90
CO ₂ purity	%	≥ 96

3. Technologies

The real potential of each separation technology for post-combustion CO₂ capture must be evaluated consistently with its state of technical development as of today, consistently with the fact that the retro-fitting of an emitting plant is a fast implementable solution, which can compensate on the short run for the unavailability of stable, economically viable and reliable carbon free

energy sources. Keeping in mind this concept, in this study we have sensibly decided to opt for ready-to-build process designs, which resort to commercially available materials and equipment.

3.1 Absorption

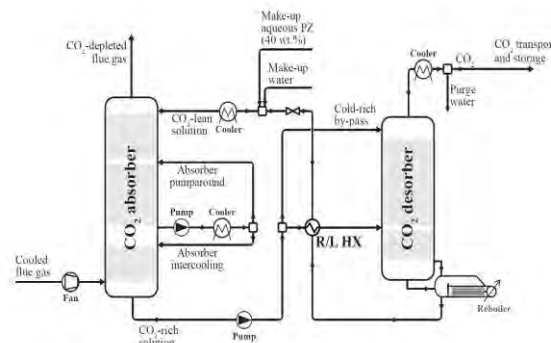


Figure 2. Process scheme of absorption capture unit.

The core of the process consists of two absorption columns, an absorber and a desorber. In the former CO₂ is removed from the flue gas by means of an aqueous solution of piperazine, a broadly studied solvent capable of fast absorption kinetics, high resistance to chemical and thermal degradation, low energy requirements for regeneration and few volatility and corrosion issues[3,12,13,14,15].

In the desorber this solvent is regenerated by providing thermal power, thus releasing a CO₂-rich gaseous product stream, which leaves the capture plant with a high water vapor content. In order to minimize the energy required for solvent regeneration, a significant share of the heat transferred to the cold loaded solvent is recovered by heat exchange with the hot lean solvent stream that exits the reboiler of the desorber, in the so-called Rich/Lean Heat Exchanger.

The most simple absorber-desorber process scheme has been also complemented with two process modifications that have been proven to improve significantly performance of the amine-based capture processes, without being capital-intensive[16]. These are: On the one hand, the intercooling of the liquid stream and the partial pump-around recycle at the absorber; on the other hand, the cold-rich bypass at the desorber. The process plant scheme is shown in Figure 2.

3.2 Adsorption

Temperature Swing Adsorption (TSA) processes use high temperatures to regenerate the sorbent after it has been saturated by the CO₂ withdrawn from the flue gas. The virtue of this solution resides in two key advantages over pressure dependent regeneration routes: 1) the possibility of using thermal energy at relatively low temperature[17,18], which is exergetically less valuable than mechanical energy and opens the way to the exploitation of recovered waste heat fluxes, and 2) the avoidance of any pre-compression of the large flue gas flow rate.

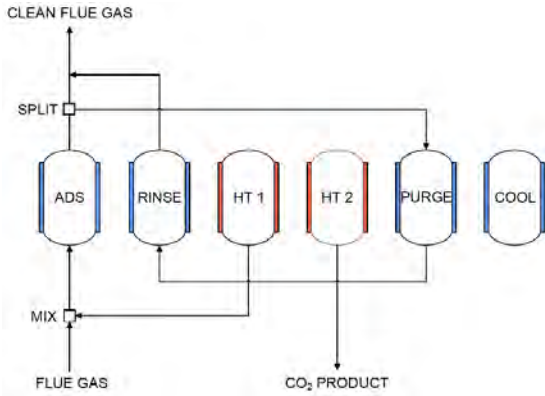


Figure 3. Process scheme of TSA cycle configuration.

The adsorption cycle configuration adopted in this study is the cycle D configuration proposed by Joss et al.[19], which improves the performance of the most standard TSA cycle by means of the introduction of a purge-rinse open loop, which recycles any CO₂ left at the end of the regeneration into the adsorption phase[20], and of a heating step split, a solution finalized at an increase in purity of the CO₂-rich product. The conceptual scheme of the adsorption cycle is displayed in Figure 3.

As regards the adsorbent, a conservative choice has been made in considering zeolite 13X, a very well known CO₂ sorbent, suitable for this type of separations and still believed to be a competitive alternative to more expensive, tailor-made materials[21,22,23]. Due to the high selectivity of zeolites towards water[24], a relevant implication of the use of this sorbent is the need for the pre-dehydration of the flue gas ahead of the separation, as previously mentioned. In this study, the flue gas is dried down to a water content of 100 ppm by means of a simple two-step TSA process, where water is adsorbed on silica gel.

3.3 Membranes

The high modularity offered by the membrane based separation systems, together with the relatively low cost of the membrane materials and the easy operation of the modules, advocate membrane separation as an interesting alternative to absorption and adsorption technologies[10,25,26,27].

In the recent years, some commercial applications specifically aimed at post-combustion CO₂ capture have

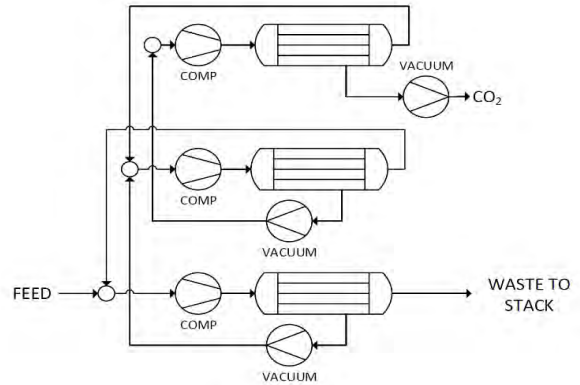


Figure 4. Process scheme of membrane capture unit.

appeared on the market. An example of this is Polaris, a polymeric membrane developed, produced and commercialized by MTR[28,29], whose favorable features place its performance close to Robenson's Upper Bound[30]. The suitability of this membrane for CO₂ capture from a binary CO₂/N₂ mixture has been also proven by several studies[29,31,32].

The capture unit analyzed in this study adopts the Polaris membrane in a series of three different separation stages, all interconnected. At each stage the retentate is recirculated back to the previous stage, while the permeate is processed further in the following one, as shown in Figure 4. At each stage, the pressure ratio required for permeation is guaranteed by the combined contribution of vacuum pumps at the permeate side and compressors at the retentate side.

As in the case of adsorption, due to the loss in performance associated with the presence of moisture, the flue gas is dehydrated ahead of the separation in a dedicated TSA unit.

4. Technical analysis

The process design of the three technologies have been optimized subject to the maximization of the unit productivity and the minimization of the exergy requirements for separation. In order to evaluate this performances, the two indicators Pr and Ex are here adopted to show the comparative results.

The definition of the exergy consumption, Ex, is common to the three technologies. Within this index

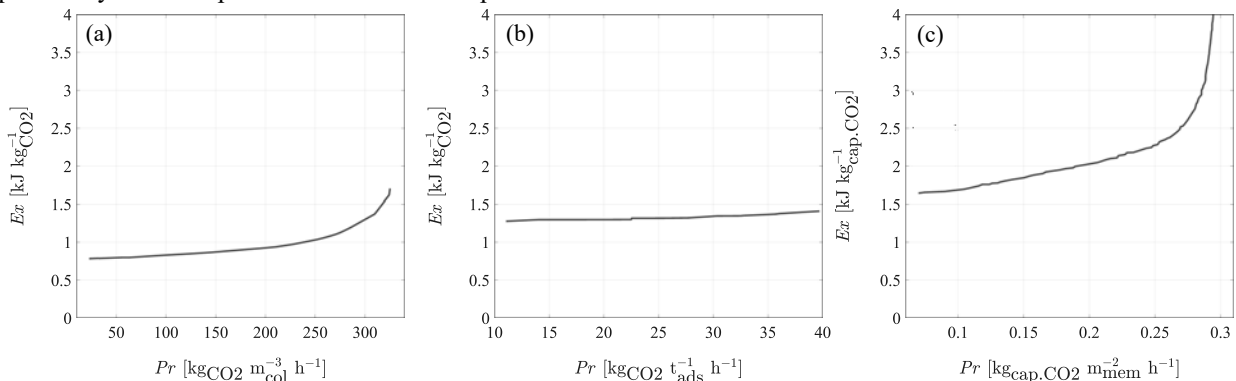


Figure 5. Comparison of the Productivity-Exergy Pareto fronts for the three post-combustion separation technologies (a) absorption, (b) adsorption, and (c) membranes.

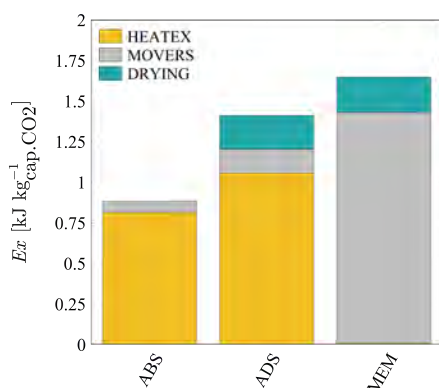


Figure 6. Breakdown of the exergy contributions of each separation technology.

different energetic contributions are considered, each of them weighted according to its exergetic content, in order to make them comparable, consistently with the concept of second principle efficiency. The total consumption is expressed in specific terms of exergy per unit CO₂ captured.

The definition of the productivity, Pr , is instead technology dependent, as it would be impossible to identify a single unit of measurement that could represent the specificity of each separation mean. In order to identify the optimum design, it is preferable to resort to an indicator which directly relates to the deployed separation material. For this reason, for each technology, the flow rate of captured CO₂ is divided here by a different denominator, that is:

- for absorption, the unit volume of vessel (including adsorber, desorber and rich/lean heat exchanger), which is also representative of the amount of piperazine solution required for the separation;
- for adsorption, the unit mass of sorbent;
- for membranes, the surface of the membrane.

The results obtained for the three technologies are displayed in figure 5. A few observations can be drawn at a first glance:

- The technologies can be ranked rather univocally in terms of exergy demand, with absorption once again proving to be the least energy intensive among the three (with a minimum at 0.8 MJ per ton of CO₂ captured), followed by adsorption, and eventually membranes.
- For all of three, the tradeoff between a lower exergy consumption and a higher plant productivity is evident, although for adsorption the curve is significantly more flat than in the other two cases, this identifying that for a TSA cycle optimizing the process configuration has a higher impact on the sizing of the plant than on the actual energy demand.

As previously mentioned, the analysis of the exergy consumption allows for the comparison of different energy sources. In fact, three technologies rely differently on heat sources and mechanical power for both regeneration and pretreatment of the flue gas (the pre-drying unit of adsorption and membranes), and clearly illustrated in Figure 6. Here the different terms forming the total exergy consumptions have been categorized according to the piece of equipment they are associated with, as follows:

- **MOVERS**, including fans, compressors and vacuum pumps for gas streams, as well as pumps for liquid streams, and thus related to the consumption of mechanical power, which in this study is assumed to be available in the form of electricity;
- **HEATEX**, including all heat exchangers, either internal or related to the provision of heat from a hot source, which in this study is identified as stream of condensing steam at the temperature required by the process;
- **DRYING**, including all pieces of equipment constituting the pre-dehydration TSA unit (only present in the case of adsorption and membrane technologies).

From a quick observation of the diagram, it is possible to infer how absorption and adsorption strictly depend on the provision of a thermal power made available by a hot source, which is the main contributor to the energy required for the regeneration of both the solvent (for the piperazine) or the sorbent (for zeolite 13X). The membrane process, instead, resorts almost completely to the mechanical power required for to move vacuum pumps and compressors.

Moreover, both adsorption and membranes pay an additional consumption fee because of the impossibility of processing a moist flue gas, which accounts for a considerable share of the total energy requirement.

5. Cost analysis

Following the same approach adopted by Hasan et al.[33,34], the cost analysis of this study is finalized to the identification of a single economic index, the Total Annualized Cost (TAC) of the capture plant, specific for unit mass of CO₂ avoided. Considering the amount of CO₂ avoided instead of the amount of CO₂ captured allows taking into account also the impact of the energy sources on the final cost of capture, consistently with the fact that the deployment of post-combustion capture units like these is realistic within a scenario where fossil fuels still play a major role in the panorama of power production.

The total annualized cost is the sum of two main contributions, i.e. the Capital Expenditures (CapEx) and Operational Expenditures (OpEx):

$$TAC = CapEx + OpEx$$

The latter includes all costs related to the utilities required for process operation (mainly the energy supplies), whereas the former is computed as the sum of the initial purchase costs of all the installed pieces of equipment. This capital investment is subject to annualization, and must be added to a further maintenance cost

$$CapEx = \phi TPC + AMC$$

with TPC being the total plant cost, AMC the annual maintenance cost, and ϕ the capital recovery factor, which relates the discount rate i and the number of annuities n (equal to the plant lifetime) and is commonly defined as

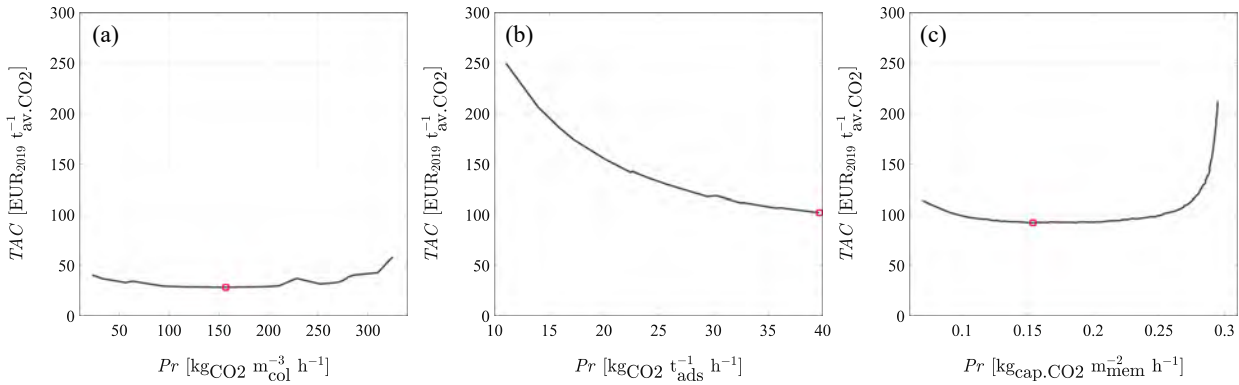


Figure 7. Comparison of Productivity-TAC curves for the three post-combustion separation technologies (a) absorption, (b) adsorption and (c) membranes. For each technology, the point corresponding to the process design associated with the minimum cost is indicated by a red square.

$$\phi = \frac{i(i+1)^n}{(i+1)^n - 1}$$

All costs are calculated according to Hasan et al. and reported in EUR referred to the year 2019. A list of assumptions made for the cost analysis is available in Table 2.

Table 2. Main parameters for the cost analysis and the calculation of the CO₂ avoided emissions.

Parameter	Unit	Value
capital recovery factor, ϕ		0.154
annual operation time	h	8000
CO ₂ footprint of steam [35]	tCO ₂ /TJ	62.3
CO ₂ footprint of electricity [36]	tCO ₂ /MWh	0.41
cost of steam	\$ ₂₀₁₉ /ton	6
cost of electricity	\$ ₂₀₁₉ /kWh	0.07
cost of piperazine solution	\$ ₂₀₁₉ /ton	6000
cost of zeolite 13X	\$ ₂₀₁₉ /ton	1300
Cost of MTR Polaris membrane	\$ ₂₀₁₉ /m ²	50

The costs have been analyzed for each technology for the chosen example case, corresponding to a daily flow rate of 10'000 tons of flue gas. The allocation of the costs is reported dividing them into a few main categories, which correspond to those already used for the analysis of the exergy consumption, apart from the additional TECH, which considers all technology specific pieces of equipment and flows.

In Figure 7 the curves of the costs corresponding to the Pareto sets of Figure 5 are displayed.

The tradeoff between CapEx and OpEx, as foreseen by the two performance indicators Ex and Pr, is evident both for the absorption and for the membrane case, where at low productivity the impact of a bigger plant size negatively affects the overall capture cost, while at high exergy consumption the cost related with the provision of mechanical and thermal power compromises the advantages offered by a more intensive exploitation of the separation mean.

The case of adsorption is, instead, different. The flat trend of the Pareto front in the productivity-exergy domain causes the cost curve to be strictly decreasing with the plant size, given that the cost related to the energy supply is rather constant along the Pareto set.

To better understand this trends, it is helpful to analyze the different shares of CapEx and OpEx for the optimum design identified along the cost curves, i.e. the process design associated with the minimum total cost. The shares for the three technologies are reported in Figure 8. The relevant role played by the cost of the equipment in the case of the TSA process is reflected in the high share of CapEx on the total cost for adsorption. The purchase cost of the adsorption columns is so high that it overcompensates the lower exergy consumption of the TSA cycle compared to the membrane process, which, in the end, is less efficient in thermodynamic terms, but nevertheless less expensive.

The advantage of the piperazine wash is, in terms of total cost, twofold. On the one hand, the low energy intensity of this capture process allows for the containment of the OpEx; on the other hand, the scalability of the absorption vessels eases the exploitation of the economies of scale that are involved in the costing of the purchased equipment, making it particularly convenient at large scale. It is to consider that in the case of both adsorption and membranes, the core separation unit (adsorption column and membrane module, respectively) cannot offer the same flexibility in terms of unit sizing. Therefore, for these two technologies a scale-out approach, rather than a scale-up one, is necessarily implemented when shifting towards larger plant sizes, resorting to large numbers of adsorption columns or membrane modules that operate in parallel. This reduces sensibly the benefit deriving from the economies of scale.

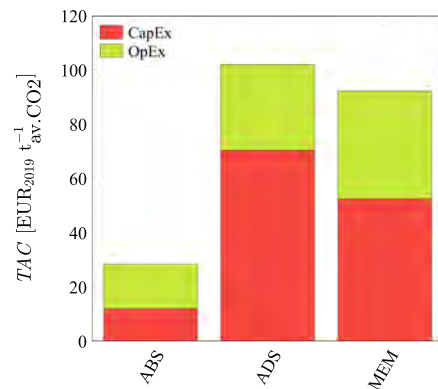


Figure 8. TAC cost for the optimum process design of each technology, highlighting the contribution of CapEx and OpEx.

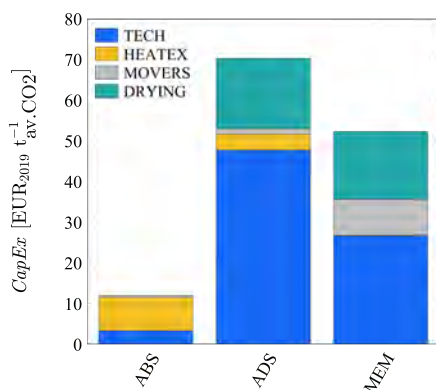


Figure 9. Cost breakdown of the CapEx associated with the optimum process design of each technology.

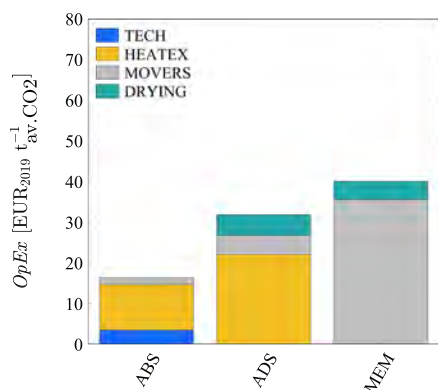


Figure 10. Cost breakdown of the OpEx associated with the optimum process design of each technology.

This effect is clearly identifiable when looking at the allocation of the cost terms for both CapEx and OpEx, as shown in Figure 9. For both adsorption and membranes, the TECH share of the cost. This includes the adsorption columns (together with their internal heat exchanging system, while HEATEX accounts for additional heat exchangers required for the conditioning of the gas streams along the recycles) and the membrane modules, for TSA and membranes respectively, and covers the largest share of the capital expenditures. Notably, the second largest share is that associated with the pre-drying unit.

As regards the operational costs, it is worth noticing that the absorption process is the only one featuring a technology related term of OpEx. It consists in the cost of the make-up flowrate of piperazine solution, made necessary by the partial volatilization and degradation of the solvent. Differently from the zeolite beads and the polymeric membrane, which require to be replaced only few times during the plant lifetime, a flowrate of solvent must be continuously fed to the amine wash plant, and is therefore accounted for as an operational cost. It accounts for about 15% of the total OpEx for absorption.

The rest of the operational cost is strictly related to the energy inputs of the systems, and thus represented by the cost of electricity for the membrane case, and of the hot steam for the other technologies.

6. Conclusions

With the present work, we have presented a comparison between three commercially ready technologies for post-combustion CO₂ capture, namely absorption, adsorption, and membranes. The process design of each of them has been firstly optimized for the capture of CO₂ out of a coal combustion flue gas, aiming at identifying a set of optimal operation designs that could express the tradeoff between low energy intensity and high plant productivity. Among this set of optima, the final process design has been elected based on total cost minimization, taking into consideration both the capital and the operational expenditures deriving from the capture process.

In conclusion, absorption has been confirmed as the least energy intensive, as well as the least expensive among the three technologies. However, the comparison has identified the rationale behind these results, highlighting how the differences in cost reflect intrinsic features of the three separation routes, in particular:

- the use of different energy sources, mainly hot utilities or mechanical power, and the consequence of their impact in terms of further CO₂ emissions, which should be accounted for by looking at the specific costs per unit CO₂ avoided;
- the technical limitations of the separation mean (solid sorbents and polymeric membranes) to perform CO₂ capture in presence of moisture;
- the technical limitations in the sizing of the pieces of equipment, which commits to the choice of a scale-out approach rather than a full scale-up when moving towards larger plant sizes, thus preventing any profit from the economies of scale.

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