

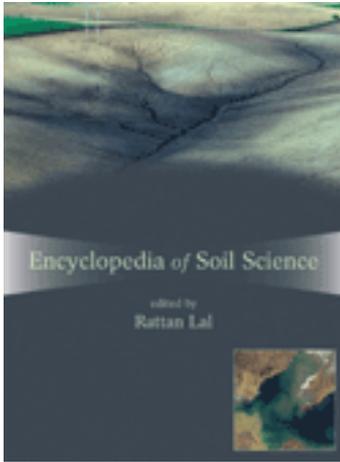
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Capture of CO₂ from Power Plants

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Abstract

An option that has been proposed to combat the buildup of greenhouse gases in the atmosphere is the capture of CO₂ at a power plant and injection of the CO₂ into a geologic formation for permanent storage, a process generally referred to as carbon capture and storage. However, because CO₂ in power plant flue gas is at a low partial pressure, current technology for CO₂ capture is expensive and could raise the cost of power by 50–100% for new pulverized coal plants, with considerably higher increases possible for existing plants retrofitted with CO₂ capture systems, when the cost of replacement power is included. This article describes current processes for CO₂ capture and some new technologies that hold promise of increasing efficiency and reducing costs.

INTRODUCTION

There is growing concern that the buildup of carbon dioxide (CO₂) in the atmosphere is contributing to global climate change. The combustion of fossil fuels, particularly coal, in power plants for the production of electricity is a major source of CO₂ emissions. A promising approach to reducing greenhouse gas emissions from fossil-fueled power plants and other large point sources is carbon capture and storage (CCS), which involves the capture of CO₂ from a large point source, transport to a storage site, and injection into a geologic formation for essentially permanent storage. The first, and most expensive, step in this chain is CO₂ capture.

CARBON DIOXIDE CAPTURE

There are three basic schemes for the capture of CO₂ from fossil-fuel-fired power plants (see Fig. 1): postcombustion capture, precombustion capture, and oxygen-combustion (oxy-combustion). In postcombustion capture, CO₂ is recovered from the flue gas produced when the fuel is burned. In precombustion capture, the fuel is decarbonized before combustion, and in oxy-combustion, the fuel is burned in an oxygen-enriched environment using pure oxygen diluted with recycled flue gas or steam to produce a flue gas consisting predominantly of CO₂ and water vapor (H₂O), from which CO₂ is recovered by condensing the water.

The advantages and disadvantages of these three options are summarized in Table 1.

POSTCOMBUSTION CAPTURE

Most fossil-fuel-fired power plants use air as the oxidant. The result is that the flue gas produced contains a large amount of nitrogen (typically about 70% for a coal-fired plant) with CO₂ concentration being in the range of 13–15%. Since flue gas is typically at about atmospheric pressure, the driving force (CO₂ partial pressure) for CO₂ capture is very low.

Because CO₂ removal from streams, such as natural gas and hydrogen, is important in the oil and gas industries, a number of processes have been commercialized for recovering CO₂ from gas streams. These processes break down into two categories: those based on physical solvents, in which the CO₂ merely dissolves in the solvent like CO₂ in seltzer water, and those based on chemical solvents, in which a weak chemical reaction occurs between the CO₂ and the solvent. The capacity of a physical solvent is a function of the partial pressure of CO₂. Physical solvents are regenerated by reducing pressure, increasing temperature, or both. Because of the low CO₂ partial pressure in flue gas, physical solvent-based processes are not feasible for postcombustion CO₂ capture.

Since chemical solvents act through a different mechanism (forming a weak chemical compound with the CO₂),

Table 1 Advantages and disadvantages of different CO₂ capture approaches.

	Advantages	Disadvantages
Postcombustion	<p>Compatible with current state-of-the-art pulverized coal-fired boilers</p> <p>Could be retrofit to the majority of existing coal-fired power plants</p>	<p>Flue gas is diluted in CO₂ and at ambient pressure</p> <p>Low CO₂ partial pressure requires use of a chemical solvent for a high capture level</p> <p>CO₂ recovery at low pressure requires a large compressor load</p> <p>The above factors result in a significant decrease in plant efficiency and output</p>
Precombustion	<p>Synthesis gas is at high pressure and has a high CO₂ concentration</p> <p>High CO₂ partial pressure results in a large driving force for CO₂ capture</p> <p>Able to use physical solvents for acid gas capture</p> <p>Recovery of CO₂ at higher pressure results in reduced compression costs</p>	<p>Applicable mainly to new plants, as few gasifier-based plants are currently in operation</p> <p>Gasifier-based power plants are not currently cost competitive, which limits applicability of precombustion CO₂ capture</p>
Oxy-combustion	<p>Very high CO₂ concentration in flue gas</p> <p>Possibility exists to retrofit and repower existing plants</p>	<p>Large cryogenic O₂ production requirement</p> <p>Cooled CO₂ recycle required to maintain temperatures within limits of materials in furnace</p> <p>Need for oxygen plant and CO₂ recycle increases auxiliary power load and decreases plant efficiency</p>

they can absorb CO₂ even when the CO₂ has a very low partial pressure. The solvent is regenerated by heating, which breaks the chemical bond and releases the absorbed CO₂. Most of these processes are based on the use of an amine as solvent. Popular solvents are monoethanol amine and diethanol amine (DEA), although other amines are sometimes used. With DEA, the reaction can be represented by:



Processes have also been developed that use a potassium carbonate solution, such as UOP's BenfieldTM process. Carbonate-based systems work by the conversion of carbonate to bicarbonate in the presence of CO₂, followed by conversion back to carbonate by heating with release of the absorbed CO₂, as indicated by the reaction:



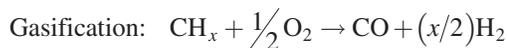
A major advantage of carbonate systems over amine-based systems is the significantly lower energy required for regeneration. Additives are sometimes included to improve absorption rate or provide other benefits.

In contrast to physical solvents, the capacity of a chemical solvent does not depend strongly on CO₂ partial pressure. Rather, it depends mainly on the concentration of the sorbent and the completeness of regeneration. Amine solvents are typically limited to concentrations of about 30% because of corrosion problems at higher concentrations. Another problem with chemical solvents is that they can form stable compounds with impurities in the flue gas and, thus, gradually lose their effectiveness. There is also a certain amount of degradation that occurs from oxygen in the flue gas. Furthermore, amine-based units have yet to be built at the scale necessary to scrub CO₂ from the flue gas of a large power plant. Some cost reductions are expected from continuing work to increase capacity and stability and reduce energy requirements.

PRECOMBUSTION CAPTURE

In precombustion capture, the fuel is decarbonized before it is burned. This typically involves gasifying the fuel, usually coal, by contacting it with a mixture of oxygen and steam to form synthesis gas (referred to as syngas), a

mixture of hydrogen (H₂) and carbon monoxide (CO), along with smaller amounts of other gases, such as H₂O, CO₂, hydrogen sulfide (H₂S), ammonia (NH₃), and methane (CH₄). Gasification is the first step in a power-producing process known as integrated gasification combined cycle (IGCC), as shown schematically in the second row of Fig. 1. The syngas is subjected to the water-gas shift (WGS) reaction to convert CO into CO₂ plus H₂. Typical reactions occurring are:



Absorbance - Antibiotics

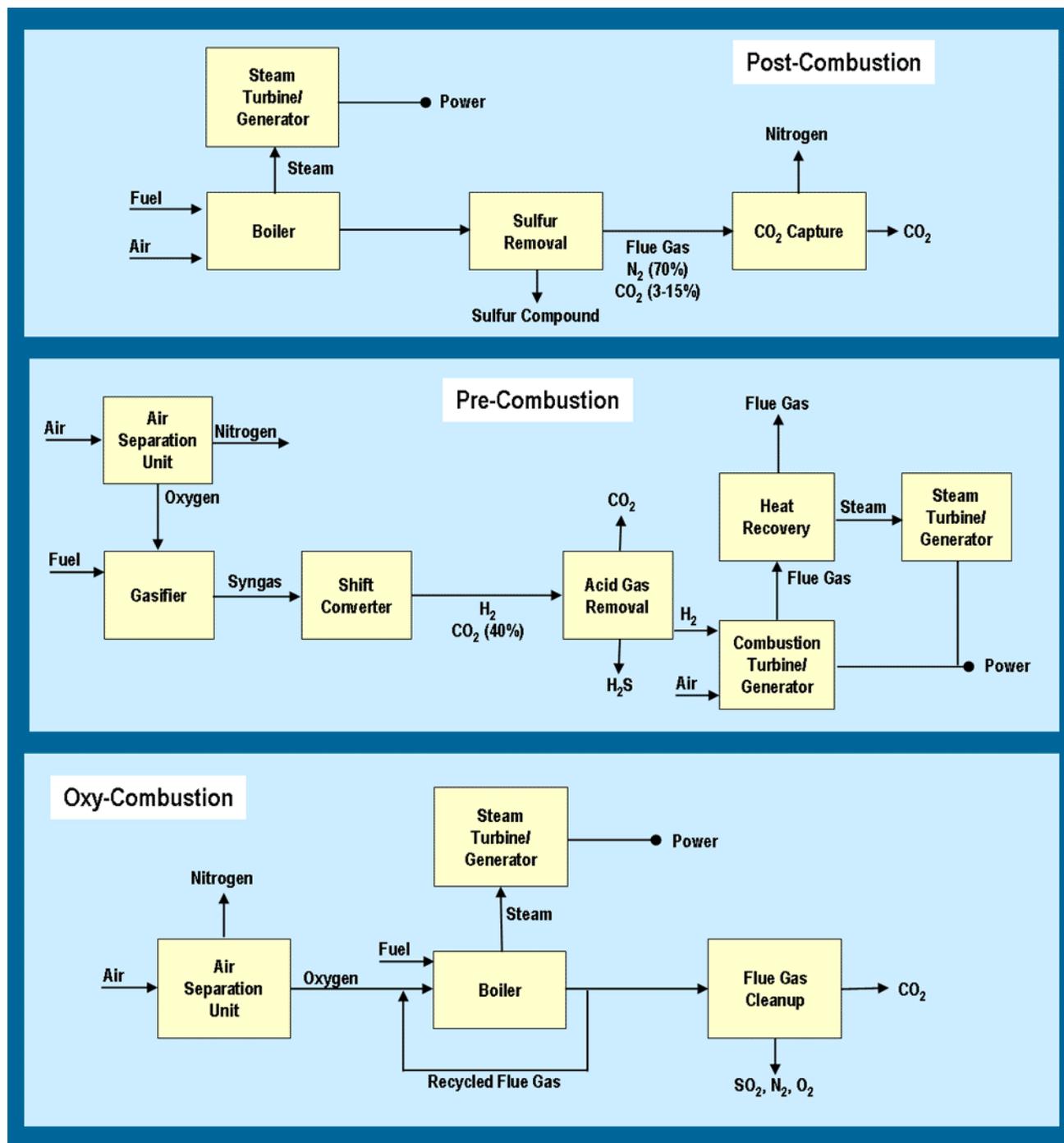


Fig. 1 Schematic showing carbon capture options.

Source: From Advances in CO₂ capture technology—The U.S. Department of Energy’s carbon sequestration program.^[1]

Other reactions also occur, but the above are the most important. After CO₂ removal, the H₂ is burned as fuel in a combustion turbine and produces only water vapor. Since H₂ has different properties than methane or CO, the combustion turbine must be redesigned to burn a high H₂ content fuel.

There are several advantages to this approach. First, gasifiers can operate at moderate pressures (500–1000 pound/inch²). This permits the use of physical solvents for CO₂ capture, rather than the more expensive chemical solvents. The most popular acid gas removal processes based on physical solvents are Rectisol®, which uses methanol as a solvent, and Selexol™, which uses a mixture of dimethyl ethers of polyethylene glycol. Second, because the gasifier runs at reducing conditions (as opposed to oxidizing conditions in a furnace), sulfur in the fuel is converted to H₂S rather than SO_x, and the nitrogen in the fuel is converted to NH₃ rather than NO_x. H₂S can be removed by the same physical solvent used for CO₂ removal, using a two-stage process, and converted to elemental sulfur, a saleable product. NH₃ can be scrubbed out with a water wash. Thus, with these systems, it is easier to remove sulfur and nitrogen in the form of H₂S and NH₃ than with systems requiring the removal of SO_x and NO_x. The biggest disadvantage of IGCC is that an oxygen plant is required, which significantly increases costs.

OXY-COMBUSTION

Oxy-combustion involves burning the fuel in an oxygen-enriched environment, where pure oxygen is diluted with recycled flue gas or steam so that the flue gas is almost entirely CO₂ and water vapor. When the water vapor is condensed, a nearly pure CO₂ stream is left. Depending on specifications and regulations, it may be necessary to remove trace contaminants, such as SO_x and NO_x, from the CO₂ before it can be injected into a pipeline for transport to a site for enhanced oil recovery or storage in a geologic formation. If CO₂ cleanup is required, costs for the oxy-combustion option will be somewhat increased.

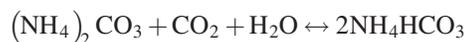
Because combustion in pure oxygen would result in furnace temperatures much higher than can be tolerated by current materials of construction, flue gas recycle is necessary, which increases costs. Another disadvantage of oxy-combustion is the requirement for an oxygen plant. In the precombustion option, only part of the fuel is burned in pure oxygen, but in oxy-combustion, all the fuel is burned in pure oxygen, so a larger oxygen plant is needed. Oxy-combustion has the potential for higher efficiency, when compared to air-fired combustion, if boiler materials that can withstand higher temperatures are developed.

EMERGING CAPTURE TECHNOLOGIES

If a new pulverized coal power plant were built incorporating current state-of-the-art CO₂ capture technology, the cost per tonne of CO₂ avoided would be about \$75, and the cost of electricity would increase by over 80%, with considerably higher increases possible for existing plants retrofitted with CO₂ capture systems, when the cost of replacement power is included. CO₂ capture is by far the most expensive step in CCS, accounting for about 75% of the cost. Therefore, for large-scale implementation of CCS to be practical, more cost-effective CO₂ capture technology is needed. Figure 2 depicts some of the emerging technologies and the approximate time frame when they may be available. Some of these technologies are discussed below.

Aqueous Ammonia

Ammonia-based systems behave similarly to amine-based systems, but have several advantages, including significantly lower heat of regeneration, higher CO₂ capacity, lack of degradation during absorption/regeneration, and low cost. The chemistry involved is as follows:



There is also the potential for the simultaneous absorption of SO_x and NO_x to form ammonium sulfate and ammonium nitrate, which can be sold as fertilizer.

A major concern with ammonia is its high volatility. This may require cooling the flue gas to the 60–80°F temperature range to reduce ammonia losses and then reheating it after CO₂ removal, which would increase costs.

Membranes

Another option under investigation is the use of various kinds of membranes to recover CO₂. As a way of separating CO₂ from flue gas, membranes have the same problem as physical solvents—a low driving force (the difference in CO₂ partial pressure on the two sides of the membrane). Since the partial pressure of CO₂ in a typical flue gas is considerably below 1 atm, a vacuum is required on the permeate side. Compressing CO₂ from subatmospheric pressure to 2000 pound/inch² (a typical pressure for geologic storage) is expensive.

Because of the higher partial pressure of CO₂, the use of membranes with precombustion capture may be more promising. Figure 3 depicts results from a polybenzimidazole (PBI) membrane under development at DOE's Los Alamos National Laboratory. This membrane has shown hydrothermal stability up to 400°C and sulfur tolerance when operating on simulated coal-derived syngas, and its performance exceeds the Robeson upper bound for H₂/CO₂ selectivity versus permeability.

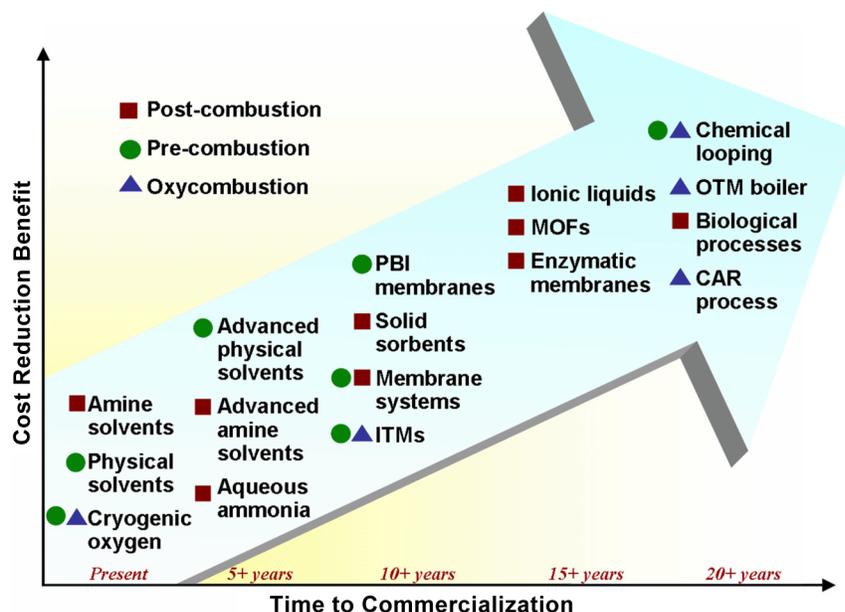


Fig. 2 Schematic showing cost reduction benefits of innovative CO₂ capture technologies versus time to commercialization. PBI, polybenzimidazole; ITM, ion transport membrane; MOF, metal organic framework; CAR, ceramic autothermal recovery; OTM, oxygen transport membrane.

Source: From *Advances in CO₂ capture technology—The U.S. Department of Energy’s carbon sequestration program*.^[1]

Solid Sorbents

Solids can act as physical sorbents (e.g., molecular sieves) or form a chemical compound, which is regenerated by heat. Many CO₂ solid sorbent processes under development tend to mirror liquid sorbent processes by being either amine or carbonate based. Amine-based systems use an amine that is stabilized by being absorbed on a porous substrate. After a CO₂ absorption cycle, the sorbent is heated to release absorbed CO₂. A carbonate system being investigated is sodium carbonate (Na₂CO₃), which reacts to form sodium bicarbonate (NaHCO₃). The bicarbonate is regenerated to the carbonate by heating.

Solid-based systems are inherently more complex than liquid-based systems because solids are more difficult to handle. Two approaches are possible: 1) moving (or fluidized) beds for the solids and 2) fixed beds with periodic gas flow switching. Because of the large volume of flue gas from a typical coal-fired power plant, equipment sizes will be large in either case.

A novel solid sorbent being investigated is metal organic frameworks (MOFs). These MOFs are a new class of hybrid material built from metal ions with well-defined coordination geometry and organic bridging ligands. They are extended structures with carefully sized cavities that can adsorb CO₂. High CO₂ capacity should be possible, and the energy required for regeneration is low.

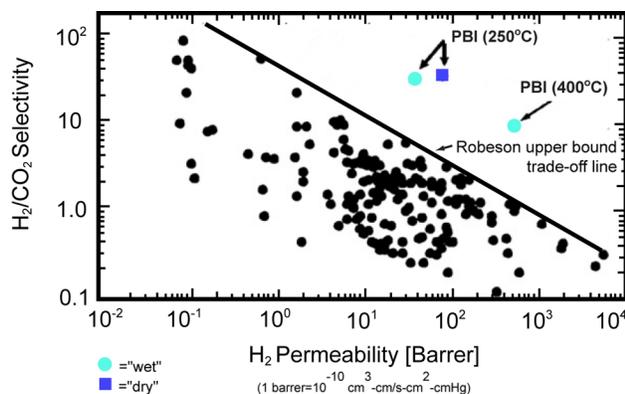


Fig. 3 Trade-off plot between H₂ permeability and H₂/CO₂ selectivity for polymers. The PBI membrane shows excellent performance.

Source: From the Los Alamos National Laboratory.^[2]

Novel Liquid Sorbents

Novel liquids being investigated as potential sorbents for CO₂ include ionic liquids (ILs). Ionic liquids are a broad category of salts, typically containing an organic cation and either an inorganic or organic anion. They can dissolve CO₂ and are stable at temperatures up to several hundred degrees centigrade, thus offering the possibility of being able to recover CO₂ from flue gas without having to cool it first. Since ILs are physical solvents, little heat is required for regeneration.

Chemical Looping

Chemical looping is a variation of oxy-combustion in which the oxygen is provided by an oxygen carrier, such as

a metal oxide, rather than an oxygen plant. The fuel is contacted by the metal oxide in a combustor, where the fuel is oxidized to produce a flue gas, which is predominantly CO₂ and water vapor, while the metal oxide is reduced. After reduction, the reduced metal is contacted with air to regenerate the oxide. This reaction is exothermic, and the hot nitrogen leaving the oxidation stage is used to raise steam for power production. CO₂ is recovered by condensing the water in the flue gas. A possible embodiment of the concept is to use two fluidized beds (similar to fluid catalytic cracking units used in petroleum refining), oxidation occurring in one vessel and reduction in the other. After heat recovery to raise steam, the oxygen-depleted air is exhausted to the atmosphere. This approach is a way to practice oxy-combustion without requiring an oxygen plant.

Improved Auxiliary Processes

If power plants are required to reduce CO₂ emissions and CCS is adopted as a CO₂ mitigation strategy, it seems likely, for the reasons discussed above, that new plants will be based on IGCC or oxy-combustion technology. However, deployment of these technologies must overcome high capital costs that affect commercial competitiveness and the ability to raise capital. One way to improve efficiency and economics for IGCC and oxy-combustion is to develop improved auxiliaries, such as air separation and WGS technology. One possibility is the use of tubular membranes. For example, tubes made of an oxygen transport membrane could be inserted into a furnace. When air is blown through the tubes, oxygen diffuses into the furnace and combusts the fuel, thus achieving oxy-combustion without the need for a cryogenic air separation unit.

Catalyst-filled membrane tubes could be used in a WGS reactor to remove one of the products (H₂ or CO₂) from the reaction, thus allowing complete reaction in one reactor, instead of requiring multiple reactors operating at successively lower temperatures.

Another area where improvements are possible is compression of CO₂. Studies are underway to determine the

most efficient compressor design for compressing CO₂ to about 2000 pound/inch², including looking at Ramjet technology. The option of compression to an intermediate pressure, followed by liquefaction and pumping to 2000 pound/inch², is also being evaluated.

CONCLUSION

Carbon capture and storage is a promising technology for reducing CO₂ emissions from fossil-fuel-fired power plants. However, current state-of-the-art technology is expensive and significantly reduces plant efficiency. Furthermore, this technology has not been implemented at the scale necessary to treat the flue gas from a large power plant. Thus, if CCS is to have an impact on mitigating global climate change effects, improved capture and auxiliary processes will need to be developed. Further information on CO₂ capture is provided in the references.^[1,3-5]

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