

# A Review on Nano-Based Technologies for Decarbonization of Chemical Industrial Processes

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Decarbonization of chemical industrial processes is a critical initiative aimed at reducing carbon emissions in the industrial sector. Decarbonization strategies encompass a spectrum of approaches.

As industries strive to meet climate goals of the Paris agreement, decarbonization plays a pivotal role in achieving a more sustainable and eco-friendly future. In the case of the hard-to-abate industries where the production of the carbon dioxide is intrinsic in the production cycle, nanotechnology-based technologies and the use of nanoparticles for exhaust gas adsorption are gaining more and more importance in this field in the last period. The present work will critically examine some of the most promising nano-based technologies for the decarbonization of hard to abate sectors.

## 1. Introduction

As global economy rapidly expands and energy consumption increases, world is confronted with the growing dual difficulties of meeting both its energy supply and demand needs. In recent times, there has been a concerning rise in the levels of CO<sub>2</sub> emissions. Consequently, it appears imperative to take measures to decrease greenhouse gas emissions (Trinca, Rispoli, et al., 2023). According to the Paris Agreement, adopted by 196 parties in 2015, to achieve the decarbonization goals and to accelerate the transition to green energy, the main focus in the short term has to be on the current industrial system. The industrial sector is the most challenging and expensive sector to decarbonize adopting the available technologies.

To diminish the carbon footprint generated by industries, it is essential to discontinue the reliance on fossil fuels as the primary energy source (Segneri et al., 2022). The most straightforward approach to achieve this is by adopting alternative energy sources such as non-fossil fuel-derived hydrogen, the thermal treatment of waste to support a circular economy and zero-waste practices, while recovering the energy contained in the waste itself, and the utilization of e-fuels (Colelli et al., 2023).

The hardest part of this transition is due to the fact that CO<sub>2</sub> emissions in the hard-to-abate industries, such as cement, steel and chemical, it is not only caused by the use of fossil fuels, but also by the fact that the generation of CO<sub>2</sub> is intrinsic in the production processes of various commodities (such as cement, lime, steel, glass etc.). The first step towards decarbonization of this sector is the replacement of the main source of CO<sub>2</sub> emission in the production cycle, as an example, in the cement production the calcium carbonate (CaCO<sub>3</sub>) is the main raw material in the production of the clinker, but it is also responsible for the majority of the CO<sub>2</sub> emission of the production site (about 60 % of the total emission); this has resulted in the need to replace limestone, clays, and shale rocks, either completely or partially, with various alternative materials (Rosa et al., 2023). These alternatives primarily consist of calcium-based substances, but also include silicates, aluminates, and iron sourced from different industrial processes.

A net zero-emission energy systems requires significant changes in the way energy is produced and used, which can only be achieved through a broad set of technologies (Falana et al., 2024). Carbon Capture, Utilization, and Storage (CCUS) is the only set of technologies that can directly reduce emissions in key sectors, as well as remove CO<sub>2</sub>, without a direct modification of the process itself. In fact, the complete

retrofitting of an existing plant, in particular in those sectors (hard-to-abate) may be the real bottleneck of all decarbonization processes. Usually, the CCS and CCU technologies act in the downstream of the process, without leading to any modification of the core (this is true for classical post and pre-combustion capture technologies such as physical and chemical absorption, physical and chemical adsorption, whereas it is not true in the case of oxycombustion or integrated calcium looping etc.).

This paper aims to give a brief notion about the actual CCS technologies used and then provide a systematic review of the nanofluids, in particularly amine-based, water-based, and methanol-based nanofluids, as potential enhancers for carbon capture processes.

## 2. State of the art

The CCS method is based on the separation of CO<sub>2</sub> from a flue gas, biogas or a syngas and its transfer, after compression and liquefaction, by pipeline to an onshore or offshore storage location (Trinca, Bassano, et al., 2023). The most important CCS technologies that are being already used are: post-combustion, oxy-combustion and pre-combustion. The post-combustion capture (PCC) is a technology that capture CO<sub>2</sub> from the flue gas of a plant. PCC has many requirements that can affect both the CapEx (Capital Expenditures) and OpEx (Operational Expenditures). The solvent is sensitive to impurities, so additional equipment is needed and lots of energy is required to regenerate the solvent that will be recycled to the absorption process. The main reasons for the use of this technology are the readiness of the technology at the required scale (TRL9) and the fact that the current structures are fully capable of being updated by this feature (Paltsev et al., 2021). Other type of PCC technologies are adsorption-based and membrane-based processes. They are characterized by relatively simple process schemes, they don't have flowing liquid phase, and have fewer auxiliary streams and fewer pieces of equipment, which make them cost-competitive at smaller scales. On the other hand, adsorption column components and membrane modules are normally limited in their maximum size and benefit less than the absorption processes from the economies of scale. Furthermore, the sensitivity of these technologies to impurities in the flue gas, such as water vapor, may vary depending on the material used. Oxy-combustion is a technology in which the fuel is burnt in high purity oxygen, instead of air. The exhaust gases obtained by combustion are mainly CO<sub>2</sub> and water vapor, with limited amount of SO<sub>x</sub>, NO<sub>x</sub> and particulates. The purification of these gases is a lot less expensive operation than the one in the PCC process; the high purity CO<sub>2</sub> stream can be processed for transport and storage after water condensation. The drawback of this process is the large amount of energy required for the production of high purity oxygen stream through an ASU and the necessity to operate on the combustion chamber or on the process core. In the pre-combustion capture systems, syngas fuel is purified from CO<sub>2</sub> usually by physical absorption. Since the syngas is usually obtained at higher pressure with respect to post-combustion case. A key benefit of this method is the high concentration of CO<sub>2</sub> in the output stream. However, this technology, like other technologies, may require great amount of energy for the regeneration of the absorbent used for the CO<sub>2</sub> capture, and also other energy to work at lower temperature than 0 °C in some cases. The last problem is the difficulty to update existing industrial structures with this system (Raza et al., 2019).

## 3. Nanoparticles for CO<sub>2</sub> chemical absorption

The high capital costs and substantial energy requirements associated with absorption processes have prompted extensive research into methods for improving CO<sub>2</sub> mass transfer while simultaneously reducing capital expenses (Trinca et al., 2023). Researchers commonly recommend two primary categories for enhancing absorption: active and inactive approaches. Active methods involve applying external forces, such as an external magnetic field, to the absorber system. Inactive methods, on the other hand, refer to mechanical and chemical approaches aimed at improving absorption performance (Hoseini et al., 2019). Nanofluids, a novel type of absorbent, are produced using an inactive method for the enhancement of the absorption performance by dispersing nanosized materials in base fluids. Nanofluids can be categorized based on the type of nano-additive that is added to the base solvent or based on the base solvent, the most important solvents used for nanofluids are amine solvents, distilled water and methanol (Tavakoli et al., 2022). Nanofluids are being explored in the field of carbon capture for their potential to enhance the efficiency of various processes involved in capturing and storing carbon dioxide (CO<sub>2</sub>). The unique properties of nanofluids can be advantageous in addressing some challenges associated with traditional carbon capture methods. Nanofluids, with their enhanced thermal conductivity and surface area, can potentially improve the absorption and desorption rates of CO<sub>2</sub> in solvents used for carbon capture processes. This can lead to more efficient capture and release of CO<sub>2</sub>. Also, nanofluids, by improving heat transfer efficiency, can help manage temperature changes more effectively, optimizing the performance of absorption and desorption units and consequentially they can reduce the duty needed to for the regeneration of the solvents leading to a decrease

of the OpEx of the carbon capture process. Another important characteristic of nanoparticles dispersed in the absorbent solutions is that they can enhance the stability of the absorbents, preventing their degradation over time; furthermore, functionalized nanoparticles can be designed to enhance the selectivity of absorbents, making them more specific in capturing CO<sub>2</sub> while minimizing the capture of other gases.

Due to their characteristics the nanofluids are suited to upgrade the post combustion technologies, enhancing the CO<sub>2</sub> absorption rate and reducing the energy requirement of this type of technologies. In some studies, to assess the reusability and reactivation of nanofluid, some tests were carried out by performing five cycles of CO<sub>2</sub> absorption and desorption. After the five cycles in the various tests depending on the nanoparticles typology, the adsorption capacity of the nanofluid decreased of only 4 - 8 % of its initial value, this implies that the solvents were able to retain, recover, and regenerate almost completely. It was also observed a decrease in the regeneration time of the solvent when the nanoparticles were added to the solvent (Lee et al., 2011).

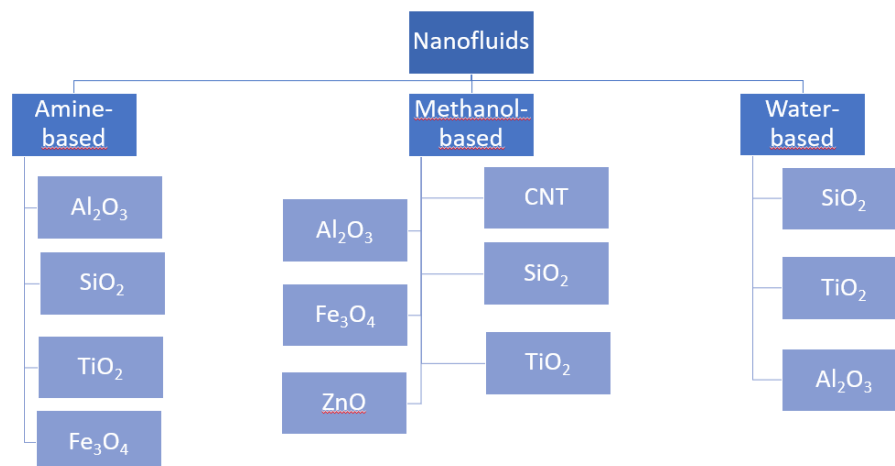


Figure1 Overview of different nanoparticles used and specific solvent

### 3.1 Amine-based nanofluids

CO<sub>2</sub> exhibits solubility in aqueous solutions containing alkanolamines (Trinca, Patrizi, et al., 2023). The effectiveness of this absorption can be enhanced by introducing PZ (Piperazine) as an activator, and similarly, additives have the potential to accelerate CO<sub>2</sub> absorption. However, due to the presence of diverse species in various amines, the chemical stability and corrosive nature of the resulting products from CO<sub>2</sub> absorption can differ. Corrosion poses challenges for the overall process, leading to a reduction in equipment lifespan. Moreover, the robust chemical bonds formed between amine solvents and CO<sub>2</sub> necessitate significant energy for the desorption process. The substantial energy requirements for solvent recovery and the corrosion issues associated with scrubbers used in CO<sub>2</sub> capture, prompted the exploration of solid sorbents as an alternative approach. Amine-modified nanomaterials have proven to be an effective solution for CO<sub>2</sub> chemical adsorption, due to their low energy consumption, chemical stability, and reversible nature. (Segneri et al., 2023).

Wang et al., (2016) conducted experimental research to explore the impact of nanoparticles on the absorption and release of CO<sub>2</sub> in aqueous MEA solutions. In the stage of diffusion-controlled mass transfer, nanoparticles demonstrate a noticeable improvement in mass transfer efficiency. Specifically, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub> nanoparticles exhibit an enhancement of over 10% in CO<sub>2</sub> absorption, with the maximum increase of the capture effect with the TiO<sub>2</sub> nanoparticles. The bubble breaking effect has the most significant role in overall mass-transfer improvement. Additionally, they investigate the enhancement of CO<sub>2</sub> desorption from rich MEA solutions with nanoparticles under various input heat fluxes. The performance ranking of the three particles is the same of the one for the increase of absorption with the TiO<sub>2</sub> leading and the SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> following. When regenerating the rich solvent from 0.4 mol CO<sub>2</sub>/mol MEA to 0.25 mol CO<sub>2</sub>/mol MEA, the solution containing TiO<sub>2</sub> nanoparticles demonstrates a time savings of approximately 42% in the desorption process.

Jiang et al., (2014), tested two different amine solvent, made of MEA and MDEA; from this study it is observable that the addition of the majority of nanoparticles can markedly enhance the CO<sub>2</sub> absorption rate. It is also shown that the most efficient nanoparticle is the TiO<sub>2</sub>. It, also, can be highlighted that the higher absorption rate is obtained with the MEA and not with the MDEA nanofluids, this is due to the higher reactivity that leads to the higher absorption rate of the base solvent, but it can be observed a higher increase in

absorption rate from the base solvent in the case of MDEA, which indicates the chemical reaction is an important factor and the enhancement effect becomes weak with the increase in the chemical reaction rate. Komati & Suresh, (2008) highlighted that with the use of a nanofluid with a magnetic nanoparticle, such as  $\text{Fe}_3\text{O}_4$ , dissolved in it, its absorption rate can be enhanced with the application of a magnetic field; results showed that with the introduction of 0.39 vol% of nanoparticles, it can be obtained an increase in absorption capacity of 92.8 % compared to the base fluid alone, this is possible with the use of nanoparticles with a diameter inferior of 15 nm, in this way the single nanoparticles can act as single-domain magnetic particles. The efficacy of  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$  nanoparticles in improving absorption was evaluated by dispersing them in three distinct base fluids: MDEA, PZ, and MEA. The mass transfer coefficient, measured on the liquid side, demonstrated a notable improvement in the absorption kinetics of the PZ absorbent following the addition of nanoparticles. It is interesting that the value of increased ratio of absorption kinetics by nanoparticle, as said before, follows the same order with solvent reaction rate with  $\text{CO}_2$ , which is PZ, then MEA and last DMEA. Results indicated that the enhancement of nanoparticle concentration reduces the absorption rate due to the solution's elasticity (Wang et al., 2016). In a different investigation, Ashrafmansouri & Nasr Esfahany, (2014) examined how the introduction of colloidal nanosilica to the 2-amino-2-methyl-1-propanol (AMP) solvent affects  $\text{CO}_2$  absorption performance within a stirred vessel. Their findings revealed that with an increase in nanoparticle concentration the absorption rate increases and the volumetric mass transfer coefficient on the liquid side decreases. In general, it can be concluded that absorption capacity increases by increasing nanoparticle concentration in the base fluid.

### 3.2 Water-based nanofluids

As a non-toxic and natural absorber, water can form physical bonds with both  $\text{CO}_2$  and  $\text{H}_2\text{S}$ . Distilled water (DW) offers certain advantages as an absorber, such as its high surface tension and suitable capacity for  $\text{CO}_2$  absorption. However, it exhibits weak performance in  $\text{CO}_2$  recovery and has a relatively low absorption rate. Consequently, numerous researchers have proposed the dispersion of nanoparticles in untreated water as a means to significantly enhance gas absorption in the resulting nanofluid (Elhaji et al., 2014).

An appropriate mathematical model was developed by Darabi et al., (2017), on the carbon dioxide adsorption by hollow fiber membranes using two different solutions of  $\text{SiO}_2$ /water and CNT/water; in this work the Brownian motion and Grazing effect were considered as the main reasons of mass-transfer enhancement. In these conditions, an increase of 32 % in the  $\text{CO}_2$  absorption rate was observed using a nanofluid with a 0.05 wt % of CNT nanoparticles, instead the increase with the use of the silica nanoparticles was only of the 16 % in the  $\text{CO}_2$  absorption rate. The reason is the lower adsorption capacity of  $\text{SiO}_2$  nanoparticles compared to CNT nanoparticles. In their work Samadi et al., (2014), studied the mass transfer performance of  $\text{CO}_2$  absorption in a wetted-wall column using various nanofluids, made of water and different nanoparticles, these studies were conducted using magnetic nanoparticles, in fact part of the tests were carried out under the effect of a magnetic field. Through this study they highlighted that the increase in liquid flow rate led to higher mass transfer flux and mass transfer coefficient and that the formation of small eddies, due to the elevated liquid flow rate, significantly improved mass transfer characteristics through their rapid movements. Adding  $\gamma\text{-Al}_2\text{O}_3$  nanoparticles to water enhanced absorption performance, and adding greater nanoparticles content in the nanofluid result in improved absorption. For instance, using  $\text{Al}_2\text{O}_3$ /water nanofluid with 1 vol % alumina particles increased the mass transfer rate by about 40–55 % compared to the solvent without nanofluid. Similarly, the mass transfer rate increased with the volume fraction of nanoparticles in  $\text{Fe}_3\text{O}_4$ /water nanofluid. However, under all conditions, the mass transfer rates and coefficients were lower than those of the base fluid (water). The use of  $\text{TiO}_2$ /water nanofluid decreased mass transfer coefficients and mass flux. At a volume fraction of 0.05 %, the reductions in mass transfer coefficient and rate were 16% and 5%, respectively.

The impact of a magnetic field on  $\text{CO}_2$ /water absorption depended on its direction. Experiments showed that a magnetic field with the same direction as the falling film strengthened  $\text{CO}_2$ /water absorption, while a field against the direction weakened it. In the presence of a magnetic field (1.5 A electric current with the same direction as the falling film), mass transfer flux and coefficient increased by 22.35 % and 59 %, respectively, compared to experiments without a magnetic field. The effectiveness of magnetic mixing can be attributed to the direct transfer of movement energy to particles by the magnetic field. This prompt particle movement increases the relative velocity between the particle surface and the surrounding medium, enhancing absorption performance. Instead, Haghtalab et al., (2015) studied the absorption rate of  $\text{CO}_2$  in pure water and the absorption rate enhancement in nanofluid composed of water and nanoparticles of  $\text{SiO}_2$  or  $\text{ZnO}$ . They highlighted that using a 0.1 wt % of  $\text{ZnO}$  nanofluids presented a higher absorption rate enhancement than the  $\text{SiO}_2$  nanofluids at the same weight of nanoparticles, for this nanofluids composition the absorption enhancement is respectively 14 % for the zinc monoxide and 7 % for the silica dioxide. Furthermore, they measured the different solubility of  $\text{CO}_2$  in the  $\text{ZnO}$  nanofluids for a range of composition of the nanofluid itself,

the percentage in weight of the ZnO ranged from 0.05 to 1 wt %, and they obtained that the solubility was enhanced by increasing ZnO weight percent, in the same temperature and pressure conditions.

### 3.3 Methanol-based nanofluids

Methanol-based nanofluids represent a unique category of absorbents that enhance CO<sub>2</sub> absorption in synthetic natural gas (SNG) systems. The absorbent's high selectivity, cost-effectiveness, and suitability for high-pressure natural gas streams make it a promising option for these applications. Additionally, its advantage lies in the lower temperatures required for the regeneration process compared to aqueous solutions, attributed to its smaller latent heat and lower boiling point. Consequently, non-aqueous alkanolamines emerge as favorable choices for improving CO<sub>2</sub> absorption performance. However, adherence to Henry's law of solubility dictates maintaining the absorbent temperature at approximately -20 °C to increase absorption rates, necessitating substantial energy consumption to sustain methanol at such low temperatures. Incorporating nanoparticles into the base fluids proves to be an effective strategy for enhancing CO<sub>2</sub> absorption in this context (Budzianowski, 2017). Lee et al., (2011) in their work obtained an evaluation of the dispersion stability for Al<sub>2</sub>O<sub>3</sub>/methanol and SiO<sub>2</sub>/methanol nanofluids at different concentrations; the authors discovered that the optimum concentration for both the nanoparticles of silica and alumina range from 0.01 to 0.05 vol % in methanol based nanofluids. It is, also, noticeable that the CO<sub>2</sub> absorption rate is enhanced up to 4.5% at 0.01 vol % of Al<sub>2</sub>O<sub>3</sub>/methanol nanofluids at 20 °C, and 5.6 % at 0.01 vol % of SiO<sub>2</sub>/methanol nanofluids at -20 °C, and also that the pH variation is closely related with the absorption enhancement by the particles dissociation. Instead, Pineda et al., (2014), worked on a rotating annular contactor with trays to test the CO<sub>2</sub> absorption rate of pure methanol and Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and TiO<sub>2</sub> methanol-based nanofluids, all tested in both co-current and counter-current; from this they highlighted that the performance of all the solvents were better in a counter-current flow system; the addition of trays enhances the absorption rate up to 9 %, 10 %, 6 % and 5 % for pure methanol, alumina, silica and titania, respectively, for counter-current flow. Furthermore, the rotation effectiveness is greater for Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> with an increase of the absorption rate for the CO<sub>2</sub> of 24.2 % and 14.4 %, respectively. The reason for the enhance of the absorption rate is the increased contact time between the liquid and the bubble due to the use of trays. From this study it is, also, discovered that SiO<sub>2</sub> nanofluids do not enhance their mass transfer any further when the liquid is put in motion.

## 4. Conclusions

The urgent challenge of contemporary society lies in the escalating problem of global climate change, primarily driven by excessive carbon dioxide emissions. This review elucidates recent progress in employing nanofluids for the absorption of CO<sub>2</sub>. From this work it can be highlighted the unique properties of nanoparticles, such as enhanced thermal conductivity, surface area, and stability, offer promising avenues for improving the efficiency of carbon capture and reducing operational costs.

Notably, the discussion on amine-based nanofluids highlights their effectiveness in chemical adsorption, with studies demonstrating significant enhancements in CO<sub>2</sub> absorption rates, particularly with the inclusion of nanoparticles like TiO<sub>2</sub>. Water-based nanofluids, despite challenges in mass transfer rates, showcase potential improvements in absorption performance, with magnetic fields proving to be a contributing factor. Meanwhile, methanol-based nanofluids present a distinctive category with applications in synthetic natural gas systems, showcasing promise in high-pressure natural gas streams.

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