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A review: Biological technologies for nitrogen monoxide abatement

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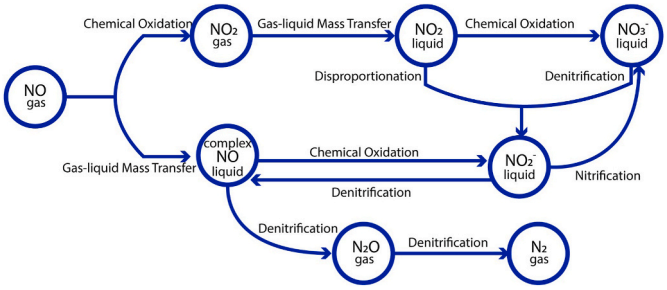
HIGHLIGHTS

- The slightly solubility of NO is a limitation for the implementation of biological treatments.
- A mass transfer vector is necessary to improve NO solubility so that biological systems can be implemented at an industrial level.
- CABR systems with $Fe(II)EDTA^{2-}$ are the most widely used biological treatments for the biological treatment of NO.
- The maximum removal capacity reported is 103.22 g NO·m-3· h^{-1} with $Fe(II)EDTA^{2-}$ in a biotrickling filter.

GRAPHICAL ABSTRACT



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ABSTRACT

Nitrogen oxides (NO_x), including nitrogen monoxide (NO) and nitrogen dioxide (NO_2), are among the most important global atmospheric pollutants because they have a negative impact on human respiratory health, animals, and the environment through the greenhouse effect and ozone layer destruction. NO_x compounds are predominantly generated by anthropogenic activities, which involve combustion processes such as energy production, transportation, and industrial activities. The most widely used alternatives for NO_x abatement on an industrial scale are selective catalytic and non-catalytic reductions; however, these alternatives have high costs when treating large air flows with low pollutant concentrations, and most of these methods generate residues that require further treatment. Therefore, biotechnologies that are normally used for wastewater treatment (based on nitrification, denitrification, anammox, microalgae, and combinations of these) are being investigated for flue gas treatment. Most of such investigations have focused on chemical absorption and biological reduction (CABR) systems using different equipment configurations, such as biofilters, rotating reactors, or membrane reactors. This review summarizes the current state of these biotechnologies available for NO_x treatment, discusses and compares the use of different microorganisms, and analyzes the experimental performance of bioreactors used for NO_x emission control, both at the laboratory scale and in industrial settings, to provide an overview of proven technical solutions and biotechnologies for NO_x treatment. Additionally, a comparative assessment of the advantages and disadvantages is performed, and special challenges for biological technologies for NO abatement are presented.

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1. Introduction

Recently, various anthropogenic activities have disturbed the natural nitrogen cycle. Prior to the arrival of the Industrial Age and the exponential growth in the world population, all reactive nitrogen (Nr) species were produced naturally from non-reactive atmospheric N_2 through natural processes, such as soil emissions and lightning (Ciais et al., 2013). The global production and industrial use of artificial nitrogen fertilizers, fuel combustion, and agriculture have led to a massive acceleration in the nitrogen cycle, generating chemical atmospheric changes owing to increased nitrogen trace gas emissions, such as nitrogen oxides (NO_x) and ammonia (NH_3). As a consequence, in recent decades, only 30–60% of Nr was a result of natural processes (Bouwman et al., 2013).

NO_x includes inorganic oxidized forms of nitrogen in the "Nr" species group (Galloway et al., 2004) that are produced by human activities, such as the combustion of fossil fuels used by transportation, the industrial sector, and power plants. In 2017, there were 7.3 Tg yr^{-1} of NO_x emissions across the 28 European Union countries. Out of these, 3.4 Tg yr^{-1} came from transportation and 2.0 Tg yr^{-1} came from the industrial use, production, and distribution of energy (EEA, 2018). Additionally, Itahashi et al. (2019) studied the variations in anthropogenic NO_x emissions from China and India between 2005 and 2016 and found that although China reached peak NO_x emissions of 29.5 Tg yr^{-1}, emissions dropped to 25.2 Tg yr^{-1} in 2016; meanwhile, NO_x emissions in India showed a continuous increase from 2005 to 2016, reaching 13.9 Tg yr^{-1} in 2016. If these trends continue, India will become the largest NO_x emitting country in the world by 2024.

The NO_x present in combustion flue gas emissions is composed of approximately 90–95% NO and 5–10% NO_2, regardless of the combustion process (Adewuyi and Sakyi, 2013). In terms of air quality, high NO_x emissions contribute to both eutrophication and acidification of ecosystems (EEA, 2017) and can lead to negative health effects, such as chronic bronchitis, asthma, and chronic obstructive pulmonary disease (WHO, 2000). As a result of these negative effects, government entities are imposing increasingly strict regulations on the emission of these pollutants.

Different strategies developed for controlling industrial NO_X emissions include the pre-treatment of feeding materials, such as fuel, oxidizer, and material being heated, and modifications in combustion processes, both of which serve as preventive measures to minimize NO_x emissions (Baukal, 2004). Nonetheless, owing to the tightening of air quality laws and high concentrations of NO_x that may be generated by combustion processes, it is necessary to implement post-treatment or end-of-pipe techniques in which NO_x is eliminated from flue gases after its formation in the combustion chamber (Baukal, 2005).

Currently, the dominant physicochemical post-treatment technologies used to control NO_x emissions from combustion gases are selective non catalytic reduction (SNCR), selective catalytic reduction (SCR) (European Commission, 2013), wet and dry scrubbing (Wen et al., 2019), and adsorption (Abdulrasheed et al., 2018). Some of these techniques have drawbacks, such as high operational or capital costs and environmental impacts, because of the abundance of hazardous wastes (secondary pollutants) that they generate, which cannot be used to create valuable products (Mladenović et al., 2018; Qie et al., 2019).

Biological NO_x treatments are now seen as alternatives to traditional physicochemical technologies because they are cost-effective and generate less secondary pollutants, making them more environmentally sustainable. Biofiltration is the most widely studied biological technology for NO_x treatment. This technique is based on the application of nitrification and denitrification.

When biofiltration is applied for NO removal, which has very low solubility in water, relatively high residence times (>1 min) are required to obtain good removal efficiencies, resulting in high reactor volumes (Jin et al., 2005). As such, several alternatives are being developed to enhance NO mass transfer and optimize bioreactor performance.

One bio-based alternative is the chemical absorption and biological reduction (CABR) process, which includes a stage prior to the biological reduction of NO, which is a chemical absorption or complexation step that occurs through the use of either a mass transfer vector or a chelating agent (van der Maas et al., 2004). Technologies such as membrane biological reactors (MBR), which are commonly used in water treatment, have also been implemented (Min et al., 2002). In addition to changing the types of reactors and processes, researchers have investigated various bio-based alternatives, including microalgae, which use NO as a nitrogen source (Qie et al., 2019), and anaerobic ammonium oxidizing bacteria (Anammox®), which can use NO as an electron acceptor and are not inhibited at high NO concentrations (Wang et al., 2018).

This review summarizes the biotechnological alternatives developed for NO reductions in combustion emissions to obtain a global vision of the current state-of-the-art and biotechnological prospects in the field of biological NO abatement. The different operating conditions and experimental efficiencies of the different investigations were analyzed, both at the laboratory pilot plant scales, to provide a general description of the technical solutions tested, as well as discuss and compare the use of the different microorganisms involved. A critical evaluation of the technological processes and the main limitations of the evaluated technologies were included.

2. Chemistry of NO_X and NO properties

There are several species of NO_X with different oxidation states in the environment, including N_2O, NO, NO_2, N_2O_3, N_2O_4, NO_3, and N_2O_5 (Skalska et al., 2010); however the abbreviation NO_X is generally related to NO and NO_2.

The oxidation and absorption mechanisms were described by Thomas and Vanderschnren (2000), and are shown in Fig. 1. In the gas phase, NO can be oxidized to NO_2 in the presence of oxygen. Other species, such as N_2O_3 and N_2O_4, can also be produced at equilibrium. In the liquid phase, species such as NO_2, N_2O_3, and N_2O_4 continuously and irreversibly react to form nitrous and nitric acids when in the presence of water.

The low Henry's constant value of NO makes it slightly soluble and almost nonreactive in water. The physicochemical properties of NO are shown in Fig. 2.

3. Conventional biological techniques

The first studies on using biological systems for NO treatment were conducted using nitrification- and denitrification-based systems, primarily in conventional biofilters, biotrickling filters (BTF), rotating drum biofilters (RDB) and membrane bioreactors (MBR) (Niu and Leung, 2010).

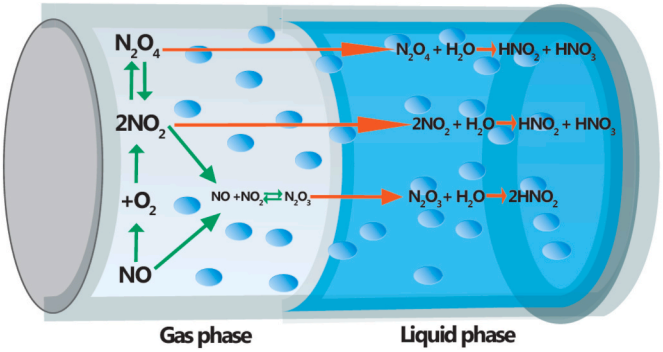

Fig. 1. Mechanisms of NOx absorption in water. Adapted from Thomas and Vanderschnren (2000).

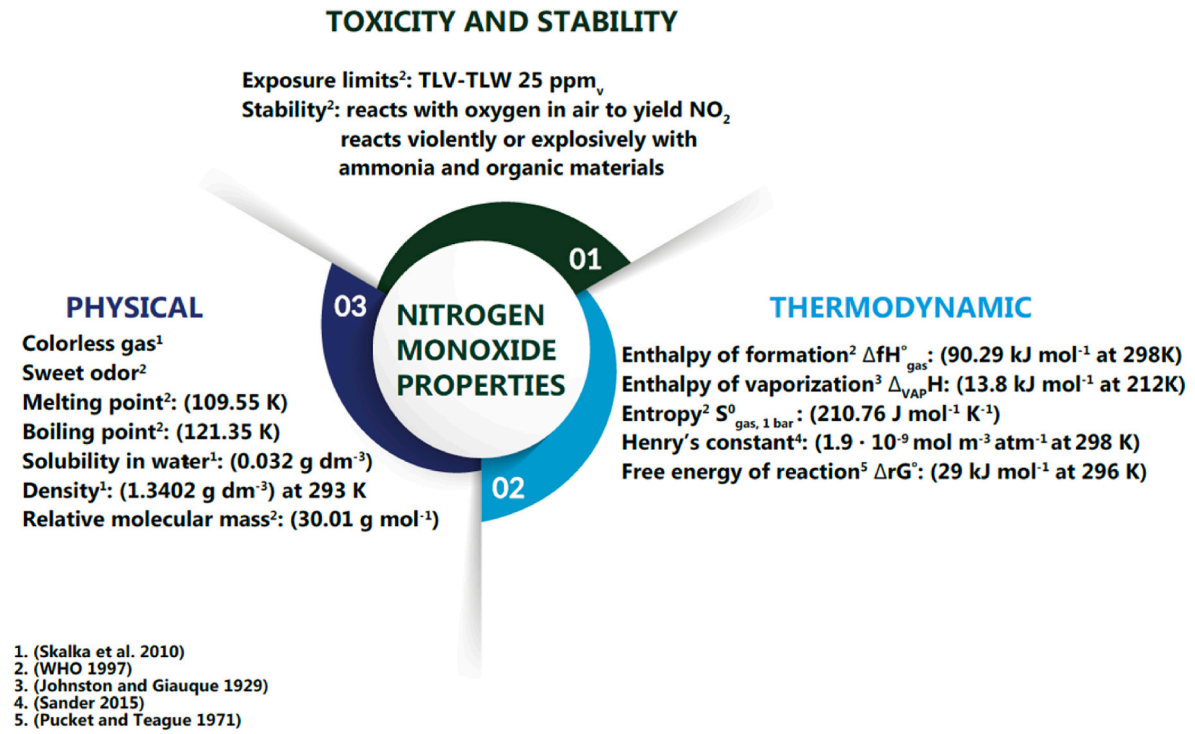

Fig. 2. NO physical, chemical, and thermodynamic properties.
(Skalska et al., 2010) (Johnston and Giauque, 1929)(Sander, 2015)(Puckett and Teague, 1971) (World Health Organization, 1997)

3.1. Nitrification process

The limitations resulting from the low solubility of NO have been studied through nitrification (Eq. (1)) using biological processes, with the aim of shifting the G–L equilibrium. Nitrification has the disadvantage of producing nitrite-(NO_2^-)- and nitrate-(NO_3^-)-rich aqueous effluents that require further treatment; therefore, it is not commonly used for denitrification.

$$NO_{(g)} + 0.75O_{2(g)} + 0.5H_2O_{(l)} \xrightarrow{\text{microorganism}} NO_{3\ (aq)}^- + H_{(aq)}^+ \quad (1)$$

Furthermore, NO biofiltration through nitrification has been tested in conventional biofilters (Okuno et al., 2000), biotrickling filters (BTF) (Chen and Ma, 2006) and hollow-fiber membrane bioreactors (HFMB) (Min et al., 2002).

In the design of bioreactors, the gas–liquid contact time is an important parameter related to the gas flow rate and reactor volume. Chen et al. (2006a) studied the empty bed retention time (EBRT) effect in a BTF running with EBRTs of 0.5, 1, 2, 3, 4, 5, and 6 min and found a 60% increase in removal efficiency (RE) with a 2 min EBRT and a slow 73% increase with a 6 min EBRT. Notably, higher EBRTs could increase investment and operating costs (higher system volumes), which would be difficult to afford on an industrial scale.

The influence of the NO inlet concentration in a BTF was studied by Chen and Ma (2006), who established that when the NO concentration was less than 100 ppm_v, biotic elimination was the primary NO abatement process, and when the concentration exceeded 1000 ppm_v, abiotic elimination, that is, NO oxidation, occurred both in the gas and liquid phases.

In wastewater nitrification processes, temperature is also important to improve system efficiency. Nevertheless, in the case of NO, the mass transfer rate changes are relatively small at high temperatures because of its low solubility. For example, Min et al. (2002) operated an MBR using synthetic combustion gas and found that the removal rate remained stable between 0.12 and 0.14 g m^{-2} d^{-1}, with an RE of 69%–73% when temperature was raised from 20 to 55 °C.

Regarding the influence of the packing material, Chen et al. (2006b) tested carbon foam and lava rock in a BTF that they operated for eight months. Their aim was to improve the mass transfer of NO and promote biofilm generation for NO removal, and they found their best results when using carbon foam, which had an RE of 94% when using an EBRT of 3.5 min and a NO concentration of less than 100 ppm_v.

Research on bioreactors using nitrification has mostly focused on the use of autotrophic nitrifying bacteria (Min et al., 2002); however, no existing study has described the microbial populations present in the system.

3.2. Denitrification process

The direct conversion of NO from a gas to liquid, followed by biological denitrification, has been widely studied in different reactor types. In the early 1990s, the first reactors to be used were the serial batch type, which was meant to determine the feasibility of integrating desulfurization and NO removal (Lee and Sublette, 1990, 1991). NO reduction was later studied in conventional biofilters (Apel and Turick, 1993; Lee et al., 2001) and BTFs (Jiang et al., 2009; Niu et al., 2014).

Heterotrophic denitrification requires the addition of carbon sources, such as organic compounds, which function as electron donors and are used for cell growth (Rahimi et al., 2020). The most widely used of these carbon sources is glucose (Wei et al., 2016b); however, some studies have also used lactate (Lee and Sublette, 1991), dextrose (Barnes et al., 1995), acetate (Flanagan et al., 2002), sodium succinate (Yang et al., 2012), and aromatic compounds such as toluene (Du Plessis et al., 1998).

Conventional bioreactors have limited NO gas-to-liquid conversion rates and require long gas residence times. Additionally, conventional biofilters and biotrickling filters present significant drawbacks because of the uneven distribution of nutrients in the reactor and the difficulty in controlling media clogging (Chen et al., 2009b). As a result, various researchers have investigated the suitability of other types of reactors. For example, Wang et al. (2006) used rotating drum biofilters (RDBs) and were able to achieve 98% RE with a NO input concentration of 529

ppm_v at an EBRT of 65 s. Furthermore, in terms of mass transfer, jet-loop bioreactors (JLBR) create more favorable conditions than conventional biofilters, yielding NO REs between 81% and 94% at NO concentrations of 500–3000 ppm_v (Durmazpinar et al., 2014).

Recently, the most widely used configuration in NO denitrification has been membrane bioreactors (MBRs), which provide a large gas-liquid surface to improve mass transfer characteristics (Zhang et al., 2013). The most studied MBR configurations include thermophilic reactors (Razaviarani et al., 2019; Wei et al., 2016a) and catalytic membrane reactors (CMRs) (Wei et al., 2016b, 2019).

One parameter that affect microbial growth and activity in bioreactors is pH. Zhang et al. (2013) studied the effect of pH on an MBR by changing the operation from a neutral pH (6–8) to a more alkaline pH (8–9). Subsequently, the elimination capacity (EC) decreased from 702 to 610 mg m^{-3} d^{-1}, suggesting that denitrifying bacteria are sensitive to pH variations, and that their highest performance occurs between neutrality and alkalescence.

In most studies, bioreactors are operated at room temperature (Razaviarani et al., 2019; Wei et al., 2016b); however, flue gases are usually emitted at high temperatures, which is why Wei et al. (2019) studied a thermophilic nitrification/denitrification MBR system between 45 and 60 °C. An efficient removal of NO could be obtained under these conditions as the bacteria acclimated to the hot flue gas and NO, reaching a maximum RE and EC of 95% and 1166 g m^{-3} h^{-1}, respectively, at 60 °C.

In biological processes, RE and EC are generally significantly improved when the EBRT is increased. This was confirmed by Chen et al. (2009a) when operating an RDB using various EBRT between 20 and 120 s, reaching a NO RE of 86% at inlet concentrations between 81.5 and 163 ppm_v and maximum EBRT.

In MBR, RT and inlet NO concentrations are also linked to RE and EC. Wei et al. (2016a) studied the influence of RT on an MBR and found that when the RT was increased from 24.5 to 49.0 s, the RE raised from 94 to 98% since increasing the RT provided enough time for NO to convert from a gas to a biofilm through the membrane, making it possible for the bacteria to degrade. Furthermore, they showed that both RE and EC decrease when there is an increase in the inlet NO concentration or a shortening of the gas residence time. In another study, Wei et al. (2019) increased the inlet concentration in an MBR from 144 to 173 g m^{-3} h^{-1} with an RT of 5 s, resulting in the NO RE decreasing from 94 to 75%, after which the bacteria adapted and recovered, and the RE increased up to 80%.

MBRs for waste gas treatment present two primary issues that reduce bioreactor efficiency, that is, excessive biofilm growth (clogging) and membrane fouling, which reduce substrate transfer (Kennes et al., 2009). Extracellular polymeric substances (EPS) and soluble microbial products (SMP) induce fouling and clogging in MBRs (Rosenberger et al., 2006). To reduce these problems, MBRs modified with new nanomaterials, coatings, and chemical grafting are currently being developed (Qin et al., 2018). For example, Wei et al. (2016b) used a catalytic MBR to reduce EPS formation and facilitate long-term operation. Additionally, Ergas (2001) reported that biofouling problems are usually greater in microporous MBRs than in dense MBRs.

Denitrification commonly occurs under anoxic conditions; therefore, oxygen is important to this process. Flue gases have oxygen concentrations of 2–8%, and the effect of O_2 concentration on NO removal has been investigated in various studies. For example, Yang et al. (2007) tested an oxygen range of 0–6% in a BTF with an inlet NO concentration of 200 ppm_v and flow rate of 30 L h^{-1}. Under anaerobic conditions the NO RE was as high as 99%, in contrast to the 55% obtained using an O_2 concentration of 6%. Similarly, negative effects were observed in another study (Niu et al., 2014) using a BTF that reached a maximum NO RE of 99% when no O_2 was present and the NO inlet concentration was 100 ppm_v at 40 °C. Recent research has studied the potential applications of aerobic denitrification-inoculating oxygen-resistant microorganisms. Zheng et al. (2016) operated a BTF under O_2 concentrations ranging from 0 to 100% and different inlet NO concentrations, reaching an RE of 92–97% in steady state. Additionally, in the latest MBR research, oxygen is present during the operating conditions, and a combination of nitrification and denitrification is presented. For example, Wei et al. (2016a) noted that, in some MBRs combining nitrification and aerobic denitrification, the effect of O_2 is not a critical operational parameter.

The effect of packing materials on denitrification has received little attention. Flanagan et al. (2002) tested three types of packing materials, that is, compost, perlite, and microporous ceramic material, and their influence on NO RE, bed pressure drops, and EBRT in a conventional biofilter. Although materials such as pearlite and microporous ceramic materials were found to have greater long-term thermal stability and reduced back pressure, the compost reached a higher NO RE of 85% in a shorter EBRT of 71 s.

3.2.1. *Microorganism involved in denitrification systems*

Generally, three types of inoculations have been observed in denitrifying bioreactors. The first type of inoculation involves different mixed cultures from wastewater plants (Chagnot et al., 1998; Wang et al., 2006) or landfill leachate (Razaviarani et al., 2019), and the second type is pure cultures. Lee and Sublette (1990, 1991) inoculated *Thiobacillus desnitrificans,* a strict autotroph, in a facultative anaerobic culture. The last type of inoculation was enriched cultures from lab-scale bioreactors that were denitrifying at a steady state (Jiang et al., 2009).

In MBRs where nitrification and aerobic denitrification were integrated, Wei et al. (2016a) found that there was synergy between nitrifying and denitrifying bacteria, such as *Nitrospira*, *Pseudomonas*, *Burkholderia*, *Burkholderia cepacia*, *Lysobacter*, *Beta proteobacterium*, *Alcaligenes* sp., and *Bacillus* strains, which are nitrifying and denitrifying while predominating and converting nitrates (originating from NO) into nitrogen by denitrification in an MBR (Wei et al., 2016b). This is consistent with the observations of Zheng et al. (2016), in which the aerobic denitrifying bacteria *Pseudomonas putida* sp. SB1 was able to consistently remove a maximum of 94% of NO in the BTF.

Several studies have been conducted under thermophilic conditions; for example, Wei et al. (2019) found that acclimatization and formation of thermophilic nitrifying and aerobic denitrifying bacteria contributed to the removal of NO. *Burkholderiales*, *Neisseriales*, *Sphingobacteriales*, and *Bacillales* were the dominant species identified in their study.

4. Chemical absorption–biological reduction (CABR)

The addition of chelating agents and absorbents is an alternative method for enhancing NO mass transfer and reducing the limitations of NO abatement through biological technologies. This was the basis for the development of a two-stage biotechnology for the removal of NO from flue gases. This technology is commonly known as CABR (Buisman et al., 2001). In the first stage of the CABR process, NO is transferred to the liquid phase via a scrubbing process using a chelating agent to reduce mass transfer resistance, and in the second stage, NO is reduced to N_2 via several biological processes.

4.1. *NO absorption using* $Fe(II)EDTA^{2-}$

When $Fe(II)EDTA^{2-}$ is used as a chelating agent in NO complexation, it reacts to produce a reversible compound, that is, the nitrosyl complex (Eq. (3)) (Schneppensieper et al., 2001a). Using this reaction, the balance in Eqs. (2) and (3) shifted towards $NO_{(aq)}$, resulting in a higher RE during the NO complexation step.

$$NO_{(g)} \leftrightarrow NO_{(aq)} \quad \text{(Eq. 2)}$$

$$NO_{(aq)} + Fe(II)EDTA^{2-}_{(aq)} \leftrightarrow Fe(II)EDTA - NO^{2-}_{(aq)} \quad \text{(Eq. 3)}$$

In addition to the complexation reaction, another undesired reaction

can occur (Eq. (4)). $Fe(II)EDTA^{2-}$ can be oxidized due to the presence of oxygen (e.g., 2–8% v/v O_2 in flue gases) to form $Fe(III)EDTA^{-}$ (Sada et al., 1987), which does not bind to NO.

$$4Fe(II)EDTA^{2-}{}_{(aq)} + O_{2(g)} + 4H^{+}_{(aq)} \rightarrow 4Fe(III)EDTA^{-}_{(aq)} + 2H_2O_{(l)} \quad \text{(Eq. 4)}$$

4.1.1. Operational conditions in absorption studies using $Fe(II)EDTA^{2-}$

The operating conditions in the absorption stage are a determining factor because they facilitate greater complexation of NO and $Fe(II)EDTA^{2-}$, reduce the oxidation of NO, and improve the RE of NO in the system. Since the reversible reaction kinetics of Eq. (3) is of first order for NO as well as for $Fe(II)EDTA^{2-}$, quick absorption of NO occurs in the system at high $Fe(II)EDTA^{2-}$ concentrations (Gambardella et al., 2006). The theoretical molar ratio of NO to $Fe(II)EDTA^{2-}$ in the complexation reaction (Eq. (3)) is 1:1 (Hishinuma et al., 1979); however, for CABR systems to be cost-efficient, a sufficient $Fe(II)EDTA^{2-}$ concentration is required for NO complexation.

The oxidation-reduction potential (ORP) has been shown to be a good indicator for identifying oxidized or reduced species in CABR systems, especially $Fe(II)EDTA^{2-}$ oxidation in CABR systems (van der Maas et al., 2006). High ORP values lead to $Fe(II)EDTA^{2-}$ concentrations that are too low to ensure a sufficiently quick NO reduction, resulting in a low NO RE (van der Maas et al., 2005a); therefore, CABR systems should operate at an ORP below −140 mV versus Ag/AgCl at a pH of 7.0.

Winkelman et al. (2007) studied the effect of temperature in complexation reactions within a range of 25–50 °C, achieving an RE of 90% at 25 °C and a decrease in efficiency with increasing temperature since at higher temperatures the complex could be oxidized and potentially destroyed (Schneppensieper et al., 2001a). Regarding the optimal pH, Huasheng and Wenchi (1988) established that, to obtain a higher complexation efficiency, the chelating solutions should be maintained at a neutral pH.

The EBRTs in the absorption towers tended to be short (<30 s) because the reaction process (Eqs. (1) and (2)) is intrinsically instantaneous (Demmink, 2000). However, Gambardella et al. (2005) found that the NO complexation process was negatively affected in the presence of oxygen; therefore, there was a longer EBRT in the column.

Reactor selection is crucial in CABR systems to ensure optimal gas-liquid contact, mass transfer, and NO RE; therefore, several reactor types have been reported in the literature.

Nitrosyl complexes were studied in different reactors before the CABR systems were investigated. For example, Nymoen et al. (1993) tested the equilibrium of $Fe(II)EDTA - NO^{2-}$ in a bubble column operated continuously at temperatures ranging from 4 to 90 °C and inlet NO concentrations between 180 and 3000 ppm_v. Bubble column reactors were used because they allow for excellent mass transfer. Nevertheless, back-mixing can occur in both the liquid and gas phases (Kantarci et al., 2005). The gas-liquid reaction in NO complexation using $Fe(II)EDTA^{2-}$ has also been studied and characterized in laboratory-scale stirred cell reactors (Demmink, 2000; Hishinuma et al., 1979; Huasheng and Wenchi, 1988).

The absorption process in CABR systems has also been studied using equipment such as sintered glass columns (Li et al., 2006; Zhou et al., 2013b), spray columns (Gambardella et al., 2006), packed-bed countercurrent columns (Winkelman et al., 2007), and sieve-plate columns (Xia et al., 2013). Meanwhile, Gambardella et al. (2006) compared a spray tower and packed column in CABR system designs, and recommended using a spray tower instead of a packed column when the reactor is operating at high mass transfer coefficient (k_L–k_G) values.

4.2. NO absorption using alternative chelating agents or absorbents

4.2.1. Other iron(II) complexes

Studies have focused on finding other iron chelators ss an alternative to $Fe(II)EDTA^{2-}$, predominantly from aminocarboxylate complexes of the iron family (e.g., 2-[bis(carboxymethyl)amino]acetic acid (NTA), 2,2'-(methylimino)diacetic acid (MIDA), hydroxyethylethylenediaminetriacetic acid (HEDTA), and 2,3-Dimercapto-1-propanesulfonic acid (DMPS)) (Demmink et al., 1997; Schneppensieper et al., 2001b). The main reactions in the CABR system with Fe(II)-L, where L is the chelate ligand, are the same as those in the systems using $Fe(II)EDTA^{2-}$ (Eqs. (2)–(4)). The main limitation to this is that iron-chelating agents that achieve a higher absorption of NO have a high sensitivity to oxygen and reactions that are not suitable for application in CABR systems (van der Maas et al., 2006); therefore, they do not improve the operation of CABR systems. Furthermore, their performance in biological systems is unknown for some aminocarboxylate complexes because they have not been investigated.

Another alternative to EDTA is nitrilotriacetate (NTA), which is less expensive and toxic than EDTA and has a moderate sensitivity to O_2 (Chandrashekhar et al., 2015; Chandrashekhar and Pandey, 2017; Wolak and van Eldik, 2002).

CABR systems using 20 mM of $Fe(II)NTA^{2-}$ for 94 d achieved greater than 90% RE in a steady state with a NO charge of 0.24 mM h^{-1} (Chandrashekhar and Pandey, 2017), similar to that of $Fe(II)EDTA^{2-}$; however, the primary drawback was that the $Fe(II)NTA - NO^{2-}$ complex is less stable than the $Fe(II)EDTA - NO^{2-}$ complex. Initially, lower sensitivities seemed to be an advantage, but Fourier Transform Infrared Spectroscopy (FTIR) analyses showed NH_4^+ and NO_2^- accumulation, derived from the NTA degradation and NO oxidation, respectively, inhibiting the microbial reduction capacity in the bioreactor (Chandrashekhar et al., 2015), which can potentially be a disadvantage for industrial-scale applications.

Another alternative chelating agent is citrate, which is more resistant to oxidation with oxygen and less expensive than EDTA. It can also be regenerated by microorganisms through denitrifying and iron-reducing bacteria (Jeter and Ingraham, 1981; Lu et al., 2011). The optimal molar ratio of Fe(II) to citrate was 1:2, with a pH of 6.9 (Liu et al., 2012).

In citrate-using studies, absorption was performed in both one-stage CABR systems using a BTF with counter-current flow (N. Liu et al., 2012; Lu et al., 2011) and two-stage CABR systems using a sinter glass column (Xu and Chang, 2007). The NO RE using $Fe(II)Cit^{2-}$ was found to be lower than that using EDTA because of its lack of capacity to complex NO; therefore, studies have focused on using a mixture of $Fe(II)EDTA^{2-}$ and $Fe(II)Cit^{2-}$ to mitigate iron oxidation and reduce operational costs. In terms of operational costs and elimination capacity, the optimal molar ratio of $Fe(II)Cit^{2-}$ to $Fe(II)EDTA^{2-}$ is 3:1 (Liu et al., 2012; Lu et al., 2011).

The maximum NO RE reported in the literature was obtained with a mixture of $Fe(II)EDTA^{2-}$ / $Fe(II)Cit^{2-}$ and a simulated flue gas concentration of 100–500 ppm_v of NO and 100–800 ppm_v of SO_2, when an RE of 90% and an EC of 43.83 g $m^{-3}{\cdot}h^{-1}$ were obtained (Lu et al., 2011). The use of $Fe(II)Cit^{2-}$ requires the use of $Fe(II)EDTA^{2-}$ to improve its efficiency, which causes limitations in implementing industrial-scale CABR systems. Studies have shown that the reduction of $Fe(III)Cit^{-}$ can be inhibited by NO_2^- and SO_3^{2-}, as is the case in CABR systems using $Fe(II)EDTA^{2-}$ (Li et al., 2011).

4.2.2. Cobalt complexes

Cobalt-based complexes have also been investigated as NO complexation options, with the most commonly used chelating agent being hexamminecobalt(II) $Co(NH_3)_6^{2+}$, where NO is dissolved in $Co(NH_3)_6^{2+}$ solution and a nitrosyl complex is formed (Long et al., 2017). This compound is able to absorb NO and O_2 and then convert them into NO_2^- and NO_3^-. Compared to iron chelates, $Co(NH_3)_6^{2+}$ can absorb O_2 for complexation reactions in ammonia solutions, instead of creating undesired oxidation reactions (Long et al., 2005). Amino carboxylate complexes of cobalt, such as cobalt(II) ethylenediamine (Long et al., 2008), cobalt(II) diethylenetriamine (Jinchao et al., 2009), and cobalt (II) triethylenetetramine (Cai et al., 2015), have also been studied, and

the complexes have been shown to have a high NO absorption capacity.

Despite showing good NO absorption results, hexamminocobalt (II) and amino carboxylate complexes of cobalt should be operated under strongly alkaline aqueous conditions (pH > 10) and cannot be used in CABR systems because NO biological reduction processes usually occur in neutral environments. No studies have shown the biocompatibility of amino carboxylate complexes of cobalt with denitrifying microorganisms (Sun and Zhang, 2018).

Jiang et al. (2019) studied different cobalt (II) chelates of monothiol-containing ligands (water-soluble amines, alcohols, or acids) containing at least one –SH group for NO removal. Specifically, systeine, mercaptosuccinic acid, 3-mercapto-propionic acid, 2-mercaptoethanesulfonate, and 2-mercaptopropionic acid complexes showed high NO denitrification potential, while being inexpensive and useable in pilot tests; however, these compounds have not yet been studied as transfer vectors for biological systems.

Currently, cobalt complexes with amino acid ligands are a promising option for use in CABR systems, because they can simultaneously absorb NO and O_2 under neutral conditions (Jezwska-Trzebiatowska et al., 1980; Zhang et al., 2016). NO absorption was found to improve when the absorbent temperature was decreased or when the cobalt(II) histidine (CoHis) concentration was increased, which was reflected in the NO removal efficiency (Sun and Zhang, 2018). Furthermore, aerobic denitrification batch tests have been performed because of their promising results in biological reduction systems (Sun et al., 2020a).

Although CoHis is highly resistant to O_2, over time, it is oxidized to cobalt (III)-histidine by autoxidation, increasing CABR system operating costs. Moreover, Co(II) is a heavy metal, where inhibition can occur in biological processes at high concentrations of chelating agents (Sun et al., 2019b).

4.2.3. Ionic liquids (ILs) and deep eutectic solvents (DESs)

One promising solution for dealing with the low NO solubility in water is the use of solvents known as ionic liquids (ILs), which include any liquids composed of ions (Begel et al., 2015). In the last two decades, ILs have been widely studied for SO_2, H_2S, and CO_2 absorption (Anthony et al., 2005; Liu et al., 2013); however, their use for NO absorption is relatively novel, and little research has been conducted on this subject.

Both the absorption capacity and the desorption process of ILs are crucial for industrial-scale implementation. A recent study, which designed an used an IL called trihexil (tetradecyl) phosphonium phenyl sulfonate [P_{66614}] [$PhSO_3$] for NO capture, showed an absorption capacity of 6.67 mol NO mol^{-1} IL at 30 °C, while the residue generated at 80 °C was 1.96 mol NO mol^{-1} IL, making it a promising alternative for application in the NO treatment (Cao et al., 2020).

The main drawback of IL usage is the difficulty of synthesizing and purifying them, resulting in high costs that can hinder their application in the chemical industry. As a result, deep eutectic solvents (DESs) are alternatives to ILs because they have the advantage of being less expensive, have a lower viscosity while achieving similar yields to ILs (Wang et al., 2021). The difference between conventional ILs and DESs is that the latter are not entirely ionic.

DESs are more easily synthesized with high purity by mixing two or more components with a fusion point lower than that of any of their individual components (García et al., 2015). DESs are usually produced by mixing hydrogen-bond donors (HBD) and acceptors (HBA) (Carriazo et al., 2012). Over the last decade, absorption tests have been performed using NO and various DESs; for example, a recent study using EG-[TEPA]Cl (3:1) for the absorption of low NO concentrations (10%), yielded a high absorption capacity of 4.53 mol NO/mol IL at 30 °C, with a significant loss of NO absorption capacity after five desorption cycles (Sun et al., 2020b).

Regarding the application of ILs or DESs as mass transfer vectors in CABR systems, the appropriate chemical structures of these families of compounds are currently being studied to improve the absorption of NO and make it reversible. Therefore, the use of these absorbents in biological treatments is still limited since their biodegradability and toxicity is not yet known.

4.3. Biological NO reduction stage in CABR systems

4.3.1. Biological reduction of NO contained in iron chelates

The biological removal of NO contained in iron chelates ($Fe(II)L-NO^{2-}$, where L is the chelating agent) has been widely studied through biological denitrification in batch tests (Table 1). Under anoxic conditions, the complete denitrification of N_2 can be achieved. The process takes place following the conventional denitrification pathway through NO reductase (NOR) and N_2O reductase (N_2OR) enzymes in the presence of an electron donor (e.g., glucose), as follows (Hollocher, 1983):

$$12Fe(II)L-NO^{2-}_{(aq)} + C_6H_{12}O_{6(aq)} \xrightarrow{\text{microorganisms}} 12Fe(II)L^{2-}_{(aq)} + 6N_{2(g)} + H_2O_{(l)} + 6CO_{2(g)} \quad \text{(Eq. 5)}$$

When oxygen levels are above 2% v/v, part of the NO is oxidized into NO_2^-, which can be further oxidized into NO_3^- by chemical oxidation or nitrification. These compounds are then reduced to N_2 (Fig. 3) (Li et al., 2014) through denitrification. In both cases, an electron donor is required (van der Maas et al., 2003).

Tables 1 and 2 show that organic compounds, such as methanol, ethanol, and glucose, are the most studied electron donors in CABR systems. More specifically, glucose is the most widely used electron donor, with an RE NO above 90% (Chen et al., 2018; Dong et al., 2013; Zhang et al., 2018); however, it is much more expensive than ethanol or methanol. Moreover, glucose concentration and its metabolite products can affect NO RE (Dong et al., 2013).

Zhang et al. (2008b) found that $Fe(II)EDTA^{2-}$ and glucose could be simultaneously used as direct electron donors for NO denitrification. Straub et al. (2001) described that denitrification with $Fe(II)EDTA^{2-}$ as an electron donor is possible because, from a thermodynamic perspective, the $Fe(II)EDTA^{2-}$/ $Fe(III)EDTA^{-}$ system is redox-reversible and has a midpoint potential of +96 mV. These characteristics allow it to donate electrons to any redox-sensitive system with a higher redox potential (NO/N_2O = +1177 mV and N_2O/N_2 = +1352 mV at pH = 7) (Zumft, 1997). However, the stoichiometry of the reaction showed that glucose could reduce NO more effectively than $Fe(II)EDTA^{2-}$ at the same molar concentrations (Chen et al., 2016b). Furthermore, Zhang et al. (2018) carried out an economic study for the future application of CABR and concluded that the operating costs can be increased by adding iron rather than glucose.

Some studies have reported that different inorganic compounds with reducing properties, such as hydrogen and sulfur-reduced compounds, can be employed as electron donors in CABR systems (Manconi et al., 2006; Xia et al., 2014).

N_2O emissions into the atmosphere should be avoided during NO reduction in any denitrification system, including CABR systems, owing to its promotion of the greenhouse effect. Therefore, high C/N ratios and low oxygen concentrations must be maintained to prevent N_2O accumulation, which could result in high organic substrate consumption and

Table 1
Summary of studies performing batch test of microbiological reduction in a CABR with $Fe(II)EDTA^{2-}$.

Chelating agent form	Chelating agent Concentration (mM)	Electron Donor	T °C	pH	Presence of free oxygen (O_2)	Maximum Removal Efficiency of NO (%)	Maximum Reduction of $Fe(III)EDTA^{-}$ (%)	Inoculum	Reference
$Fe(II)EDTA^{2-}$	20–25	Ethanol Acetate Hydrogen Molasses	55	7–7.4	Anaerobic	20	n.a.	Anaerobic methanogenic granular sludge and denitrifying sludge from wasterwater treatment plants	van der Maas et al. (2003)
$Fe(II)EDTA-NO^{2-}$ $Fe(III)EDTA^{-}$	7	Glucose	40	6.98	Anaerobic	91.14	87.8	*Pseudomonas* sp. - *Klebsiella trevisan* sp.	Jing et al. (2004)
$Fe(II)EDTA^{2-}$	25	Ethanol Acetate Methanol Fe(II) $EDTA^{2-}$	30–55	7–7.4	Anaerobic	n.a.	n.a.	Sludge of full-scaled biological fluidized bed reactor (BFBR)	van der Maas et al. (2004)
$Fe(II)EDTA^{2-}$	10	Ethanol Fe(II) $EDTA^{2-}$	30	7	Anaerobic	n.a.	n.a.	Sludge of lab-scale denitrifying reactor – *Paracoccus denitrificans*, *Paracoccus pantotrophus* and *Paracoccus versutus*	Kumaraswamy et al. (2006)
$Fe(II)EDTA-NO^{2-}$ $Fe(III)EDTA^{-}$	3.3 6	Glucose Fe (II)$EDTA^{2-}$	40	6.8–7.2	Anaerobic	85.8	99	Pseudomonas sp. Strain DN-2	Zhang et al. (2007)
$Fe(II)EDTA^{2-}$ $Fe(III)EDTA^{-}$	0–40	Glucose Fe (II)$EDTA^{2-}$	40	5–10	Anaerobic	n.a.	87	*Escherichia coli* strain FR-2	Li et al. (2007)
$Fe(II)EDTA^{2-}$ $Fe(II)EDTA-NO^{2-}$	45	Ethanol	35	7	Anaerobic	n.a.	n.a.	Denitrifying and strictly anaerobic sludge from wastewater treatment plant	Dilmore et al. (2007)
$Fe(II)EDTA^{2-}$	2–10	Glucose	40	6.2–6.8	Anaerobic	n.a.	n.a.	Actived sludge wastewater treatment plant - *Enterobacter cloacae*	Zhang et al. (2008b)
$Fe(II)EDTA^{2-}$	2–25	Ethanol	55	7–7.4	Anaerobic	20	n.a.	Methanogenic granular sludge. denitrifying sludge and CABR sludge reactor	van der Maas et al. (2008)
$Fe(III)EDTA^{-}$	10	Glucose	30–60	3–8	Anaerobic	n.a.	93.5	Actived sludge wastewater treatment plant	Jing et al. (2012)
$Fe(III)EDTA^{-}$	3.6	glucose pyruvate lactate ethanol acetate propionate	30	7.8–7.2	Anaerobic	n.a.	92	*Paracoccus denitrificans ZGL1*	Dong et al. (2012)
$Fe(III)EDTA^{-}$ $Fe(II)EDTA-NO^{2-}$	2 4 8	Glucose	30	7	Anaerobic	n.a.	77.5	Seabed sludge - *Paracoccus Versutus* LYM	Dong et al. (2013)
$Fe(III)EDTA^{-}$	10	Glucose	40	2.9–10.3	Anaerobic	n.a.	99.2	Cultivated mixed culture *Klebsiella* sp. *FD-3*	Wang et al. (2013b)
$Fe(III)EDTA^{-}$ $Fe(II)EDTA-NO^{2-}$	0–10	Glucose	40	6.7–6.9	Anaerobic	74.1	99.6	Cultivated mixed culture *Klebsiella* sp. *FD-3*	Zhou et al. (2013a)
$Fe(III)EDTA^{-}$	25	Glucose	30	6.8–7.6	Anaerobic	56.3	n.a.	*Paracoccus denitrificans* strain ZGL1	Dong et al. (2014)
$Fe(III)EDTA^{-}$ $Fe(II)EDTA-NO^{2-}$	0–4	Glucose	40	n.a.	Anaerobic	91	99.6	Actived sludge wastewater treatment plant enrichment	Lin et al. (2014)
$Fe(II)EDTA-NO^{2-}$	5	Glucose	40–60	6.6–7.1	Anaerobic	98.7	n.a.	Denitrifying bacteria - *Anoxybacillus* sp. HA	Chen et al. (2015a)
$Fe(II)EDTA-NO^{2-}$	2–5	Glucose Fe (II)$EDTA^{2-}$	n.a.	7–7.4	Anaerobic	85	n.a.	Anaerobic sludge of a sewage treatment plant an enrichment	Chen et al. (2016b)

n.a not available.

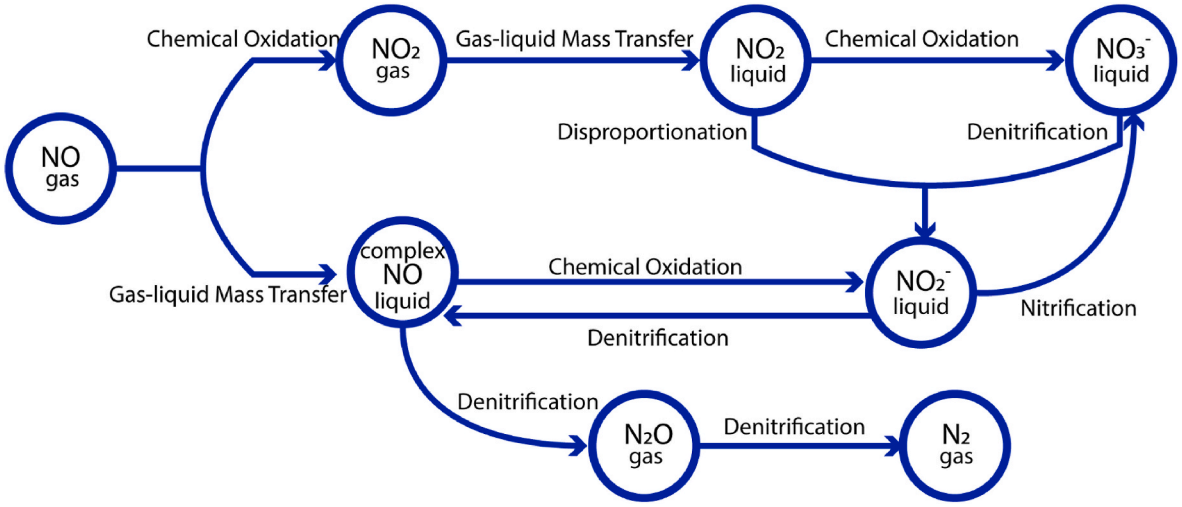

Fig. 3. NO Conversion pathways, adapted from Li et al. (2014).

increased operational costs (Chen et al., 2016b). Meanwhile, Sørensen et al. (1987) described that the use of sulfur compounds, such as sulfide (S^{2-}), as electron donors can also result in N_2O accumulation due to possible bacterial activity inhibition at concentrations above 1 μM. The same phenomenon was observed by Manconi et al. (2006) when a high concentration of SO_3^{2-} accumulated in the system and its heterotrophic reduction competed against denitrification.

Apart from the different batch tests using $Fe(II)EDTA^{2-}$ that are presented in summary Table 1, the CABR systems were also studied under several operating conditions and different inocula in continuous reactors using $Fe(II)EDTA^{2-}$, as presented in Table 2, and in continuous reactors using other chelating agents, which are summarized in Table 3.

Denitrification in one-stage CABR systems has been commonly studied in BTFs operated in countercurrent mode (Chandrashekhar et al., 2013; Dilmore et al., 2006; Lu et al., 2011) and in RDBs to prevent the operational limitations generally associated with BTFs, such as uneven nutrient distribution and packing material clogging (Chen et al., 2007). Nevertheless, the best performance was observed in two-stage CABR systems, where inhibition was reduced by the influence of oxygen on microbial activity, ensuring a steadier operation.

As flue gases generally enter the process at high temperatures, bioreactors are operated at moderate temperatures between 35 and 55 °C. Researchers have sought to find the optimal temperatures for the growth, reproduction, and metabolism of bacteria capable of reducing $Fe(II)EDTA-NO^{2-}$ and $Fe(III)EDTA^{-}$. For example, *Anoxybacillus* was found to have an optimal temperature of 55 °C yielding a NO RE of 95%, and when the temperature was raised to 60 °C, the RE NO decreased to 85% because high temperatures can lead to denaturation of nucleic acids or proteins (Chen et al., 2015a). The optimal temperature for *P. denitrificans* sp. ZGL1 and *Pseudomonas* sp. DN-2 is in the range of 30–40 °C because a lower temperature could inhibit enzyme activity (Dong et al., 2014; Zhang et al., 2007).

As can be seen in Tables 1–3, CABR systems should primarily be operated under pH conditions ranging between neutral and lightly alkaline because this is proven to be the most suitable range for denitrification, as shown by Chen et al. (2013a) in an RDB where an RE of 84% was reached with a pH of 8.0–8.5; however, when pH was increased to 9.0, RE was reduced to 70%, which affected the metabolism of the bacteria.

One major drawbacks that prevents the industrial-scale implementation of CABR systems is the influence of the inlet NO concentration. That is, at high NO concentrations, the $Fe(II)L-NO^{2-}$ formation rate exceeds the maximum microbial reduction rate and causes RE to diminish (Li et al., 2006). This was proven by Zhou et al. (2013b) using a two-stage CABR system in which a NO RE of 95% was obtained in between 122.3 and 448.3 ppm_v; however, when the NO concentration was increased to 733.5 ppm_v and 1043 ppm_v, the RE decreased to 88% and 75%, respectively.

CABR systems are strongly affected by EBRT; however, increasing the EBRT in bioreactors is not economically viable, while reducing it can cause debilitation of microbial activity and acceleration of the consumption of $Fe(II)EDTA^{2-}$, leading to an increase in operational costs and a decrease in the reaction time (Chen et al., 2018). Lu et al. (2011) showed that when the EBRT was reduced from 120 to 60 s, the NO RE decreased from 95% to 75% because the formation rate of $Fe(II)EDTA-NO^{2-}$ exceeded the treatment capacity of the microorganisms. The EBRT values varied widely in the different CABR studies; however, the majority were between 10 and 180 s.

The hydraulic residence time (HRT) is a key factor in the operation of reactors and in the microbiological reduction of NO because the energy consumption of the system can affect operational and investment costs. A wide variety of HRT values have been used in various NO reduction reactors, that is, 22.4 min (van der Maas et al., 2005b), 8 h (Chandrashekhar and Pandey, 2017; Liu et al., 2014) and 20 min (van der Maas et al., 2006). Overall, higher HRTs generally result in higher investment costs.

Regarding reactor configuration, Zhang et al. (2018) compared a two-stage CABR and a one-stage CABR and found that the primary drawback observed in a one-scale CABR was that it was impossible to maintain a stable NO RE at a higher gas flow rate, owing to the toxicity of the microbial activity of O_2 and NO. Additionally, the gas flow rate was reduced and the gas-liquid contact was diminished, resulting in higher residency times in the reactors.

4.3.2. Biological regeneration of the iron chelating agents

The regeneration process for iron-chelating agents in CABR systems occurs during denitrification (Eq. (5)) through the reduction of $Fe(III)L^{-}$, which is biologically mediated by iron-reducing bacteria,

Table 2
Summary of studies using continuous reactors for the microbiological reduction in a CABR with Fe(II)EDTA^{2-}.

Bioreactor type	Inlet NO concentration (ppm_v)	Volumetric Flow Rate (m^3h^{-1})	Electron donor	T °C	pH	Presence of free oxygen (O_2) %	EBRT (s)	Maximum Removal of Efficiency NO (%)	Elimination Capacity (Ec) ($g\ m^{-3}.h^{-1}$)	Inoculum	Reference
BTF	70–500	0.65	Ethanol	55	Upperlimit 7.6	0.8–3.3	10	80	14.45–103.22	Denitrifying and methanogenic sludge	van der Maas et al. (2005a)
Two stages	1200–1500	123	Ethanol	40–60	7–8	3–6	13	99	7.53–12.43	*Deferribacter thermophilus, Denitrovibrio acetophilus, Bacillus infernus, Bacillus simplex, Bacillus thermodenitrificans* and *Bacillus azotoformans*	Kumaraswamy et al. (2005)
Two stages/packed-bed scrubbing column + upflow bioreactor	1525	0.19	Ethanol	35	7.1	Anaerobic	805	97.9	5.15	Denitrifying and strictly anaerobic sludge from wastewater treatment plant	Dilmore et al. (2006)
Two stages/sintered glass packed column + packed with glass fiber column	100–500	0.06	Ethanol Glucose	50	6.5–7.0	0–8	n.a.	88	7.32–36.60	Actived sludge wastewater treatment plant enrichment	Li et al. (2006)
RDB	65–350	0.06–0.29	Glucose	30	5–9	Anaerobic	30–150	96.5	1.23–33.10	Activated sludge wastewater treatment plant enrichment	Chen et al. (2013a)
BTF	0–546	0.12	Glucose	45–55	6.5–7.5	0–10	90	>90	16.05	Pseudomonas sp. DN-2 and *Escherichia coli* FR-2	Liu et al. (2014)
BTF	0–546	0.12	Glucose	49.5–50.5	6.8–7.2	0–10	n.a.	90	n.a.	*Pseudomonas* sp. DN-2 and *Escherichia coli* FR-2	Li et al. (2014)
BTF	100–500	0.06	Glucose	50	6.6–6.8	3–6	180	95	1.55–7.76	Actived sludge wastewater treatment plant enrichment	Li et al. (2016a)
BTF	0–400	0.06	Glucose	50	6.8–7.0	0–12	120	99	9.7	Actived sludge of denitrifying reactor enrichment	Li et al. (2016b)
BF	163–1630	n.a	n.a	49–51	n.a	8	42	89.8	146.9	*Chelatococcus daeguensis* TAD1	Han et al. (2016)
Two stages/ Absorption column and bio bioreactor	200–800	0.06–0.36	Glucose	50	6.8–7.2	0–9	13.44–80.64	89.10	n.a	Activated sludge in a sewage treatment plant	Zhao et al. (2017)
Two stages/sieve plate column + packed-bed column	400	0.06	Glucose	50	6.8–7.2	0–12	30	>90	9.93	Iron reducing and denitrifying bacteria	Zhang et al. (2018)
RDB	122–820	n.a.	Glucose	25	7	2	90	95	n.a.	Actived sludge wastewater treatment plant enrichment	Chen et al. (2018)

n.a not available.

Table 3
Summary of studies using other chelating agents in CABR systems.

Experimental system characteristics	Carbon Source	Chelating agent	Inlet NO (ppm_v)	Volumetric flow rate (m^3h^{-1})	T °C	pH	Presence of free oxygen O_2 (%)	EBRT (s)	Maximum Removal Efficiency of NO (%)	Elimination Capacity (Ec) ($g\ m^{-3}{\cdot}h^{-1}$)	Inoculum	Reference
Continuous/ Two stages – sintered glass packed column + packed with glass fiber column	Glucose	$Fe(II)Cit^{2-}$	250–1000	0.06	55	6.5–7.0	0–10	300	59	0.14–0.58	Activated sludge of a sewage treatment plant	Xu and Chang (2007)
Batch	Glucose	$Fe(III)Cit^{-}$	n.a.	n.a.	40	6.7–6.9	Anaerobic	n.a.	n.a.	n.a.	Sludge lab scale bioreactor – *Enterococcus* sp. FR-3	Li et al. (2011)
Continuous/ BTF	Glucose	$Fe(II)Cit^{2-}/Fe(II)EDTA^{2-}$	100–800	0.048–0.12	45–55	6.7–6.9	1–5	60–150	>90	43.83	Actived sludge wastewater treatment plant	Lu et al. (2011)
Continuous/ BTF	Glucose	$Fe(II)Cit^{2-}/Fe(II)EDTA^{2-}$	0–546	0.048–0.12	45–55	3.47–8.54	0–6.5	16–39	80	86.54	Actived sludge wastewater treatment plant	Liu et al. (2012)
Batch	Ethanol	$Fe(II)EDTA^{2-}Fe(II)NTA^{2-}Fe(III)EDTA^{-}Fe(III)NTA^{-}$		n.a.	40	5–6.78	Anaerobic	n.a.	n.a.	n.a.	Sludge of a Sewage treatment plant – *Enterobacter, Citrobacter* and *Streptomyces*	Chandrashekhar et al. (2013)
Continuous/ Two stages – CSTR + packed-bed column	Ethanol	$Fe(II)NTA^{2-}$	97–245	0.003–0.0035	35–39	4.9–7.4	1–2	n.a.	87.8	0.023–0.066	Sludge of a Sewage treatment plant – *Enterobacter, Citrobacter* and *Streptomyces*	Chandrashekhar et al. (2015)
Continuous/ Two stages – CSTR + packed-bed column	Ethanol	$Fe(II)NTA^{2-}$	81–250	0.1	37	7	1–2	n.a.	>90	n.a.	Sludge of a Sewage treatment plant – *Enterobacter, Citrobacter* and *Streptomyces*	Chandrashekhar and Pandey (2017)

n.a. not available.

using an organic carbon source as an electron (Eq. (6)):

$$24Fe(III)L^{-}_{(aq)} + C_6H_{12}O_{6(aq)} + 24OH^{-}{}_{(aq)} \xrightarrow{\text{microorganisms}} 24Fe(II)L^{2-}_{(aq)} + 18H_2O_{(l)} + 6CO_{2(g)} \quad \text{(Eq. 6)}$$

Liu et al. (2014) studied glucose consumption in CABR systems and concluded that the accumulation of acetate generated from glucose fermentation inhibited the Fe(III)EDTA$^-$ reduction process.

Furthermore, some studies have also reported that iron reduction can be performed chemically through sulfur-reduced species as extracellular electron shuttles (van der Maas et al., 2005a). As such, S^{2-} can be exploited to reduce iron (Eq. (7)) (Frare et al., 2010; Wubs and Beenackers, 1994). Chen et al. (2015b) confirmed that Fe(III)EDTA$^-$ reduction is biologically driven by sulfur-oxidizing bacteria (SOB) and chemically driven by CABR systems.

$$S^{2-}_{(aq)} + 2Fe(III)EDTA^{-}{}_{(aq)} \rightarrow S^{0}_{(aq)} + 2Fe(II)EDTA^{2-}_{(aq)} \quad \text{(Eq. 7)}$$

Zhang et al. (2012) reported that O_2 levels lower than 3% inhibit iron-reducing activities, and Arnold et al. (1990) found that an increase in O_2 slows down the biological regeneration of Fe(II)EDTA^{2-}. Furthermore, pH changes in the CABR systems inhibit bacterial growth. Jing et al. (2012) studied the effect of pH on immobilized and free bacteria and found that the optimal pH was 7.0 and that a decrease in pH resulted in less reduction of Fe(III)EDTA$^-$.

The accumulation of inhibitors of iron-reducing bacteria, such as N_2O, Fe(II)EDTA – NO^{2-}, NO_2^-, NO_3^-, and sulfur compounds, such as SO_3^{2-} also negatively affects the regeneration of the chelating agent (Chen et al., 2019; Li et al., 2016a; Manconi et al., 2006; van der Maas et al., 2008).

4.3.3. Microorganisms involved in CABR systems

The performance of the CABR process relies on the capacity of the system to reduce both NO to N_2 and Fe(III)EDTA$^-$ to Fe(II)EDTA^{2-}. The success of this process is strongly related to the microorganisms involved in both the biological processes. Zhang et al. (2007) reported that organisms such as *Pseudomonas* sp. DN-2 can reduce both Fe(II)EDTA – NO^{2-} and Fe(III)EDTA$^-$ using glucose and Fe(II)EDTA^{2-} as electron donors. However, the use of other bacteria is necessary for the process to be viable because when Fe(III)EDTA$^-$ is present, a Fe(II)EDTA – NO^{2-} reduction of only 57% was achieved. Meanwhile, when Zhang et al. (2008a) used a mixture of denitrifying and iron-reducing bacteria, such as *Pseudomonas* and *Escherichia coli*, the NO RE reached 70%.

Li et al. (2016a) compared a CABR system with pure and enriched cultures of steady state oxygen-resistant denitrifying bacteria operated under the same conditions. They found that the system that used enriched cultures yielded a higher NO RE (99%) despite operating at an O_2 concentration of 6% v/v. Meanwhile, in the CABR system with pure cultures, a lower NO RE of 90% was obtained at an O_2 concentration of 3% v/v. This was likely because the enriched crops were able to adapt to extreme conditions, and therefore, a higher ER was achieved.

Other CABR systems use various inoculums, including sludge from wastewater treatment plants (Dilmore et al., 2006) and enriched cultures such as *Pseudomonas* and *Klebsiella trevisan* (Jing et al., 2004), while others are inoculated with isolated pure cultures such as *Paracoccus denitrificans* (Dong et al., 2014).

Kampf (2002) reported that immobilization, rather than microorganism suspension, led to higher efficiencies because it protected bacteria from unfavorable environments and maintained the integrity of the bacteria and a higher cell density. Furthermore, Zhou et al. (2013b) confirmed that in a two-stage CABR system, a high removal efficiency (95%) was reached due to the gradual acclimation of the immobilized bacteria in the biofilm, whereas an efficiency of 85% was reached after 5 d of continuous operation with suspended microorganisms.

In another innovative system, an Fe(III)EDTA$^-$ removal of ~ 100% was achieved by immobilizing iron-reducing bacteria using Fe_3O_4-chitosan magnetic microspheres (Jing et al., 2012). Similarly, a NO RE of 99% was achieved at 40 °C by immobilizing *Klebsiella* sp. FD-3 with Fe_3O_4 polystyrene glycidyl methacrylate magnetic porous microspheres (Xiaoyan Wang et al., 2013).

4.4. Drawbacks of CABR systems in NO abatement

Currently, there is no reported evidence that industrial-scale CABR systems have been implemented, which could be related to the instability of the chelating agent used, rather than the technology itself. For example, the exposure of Fe(II)EDTA^{2-} to sunlight for long periods has been shown to result in the destabilization of the complex (Lockhart and Blakeley, 1975; Santiago et al., 2010). Likewise, the chemical structure of chelating agents depends on operating parameters, such as a pH of 4–10 (Demmink et al., 1997) and a temperature of 25 ± 0.5 °C (Schneppensieper et al., 2002), if it cannot change the kinetics and thermodynamics of the absorption process (Gambardella et al., 2005).

The long-term operation of CABR systems is hindered predominantly by the oxidation of chelating agents, which causes a reduction in RE and the loss of total iron in the recirculation system. Therefore, the extra consumption of the chelating agent derived from the constant oxidation (Eq. (4)) of the complex to Fe(III)EDTA$^-$, therefore, leads to an increase in operational costs.

Van der Maas (2005) reported that iron precipitates in scrubber solutions suppressed the NO absorption rate because of the accumulation of these precipitates in the packing material or in the column absorption plates, which prevents good gas-liquid contact and efficient mass transfer.

One of the main disadvantages of this system is the loss of the chelating agent due to its degradation. Li et al. (2016b) quantified iron loss in a BTF over 72 h and found a 44% iron precipitation. X-ray photoelectron spectroscopy (XPS) revealed that this resulted from the formation of $Fe(OH)_3$ since chemical or biological degradation of EDTA facilitates iron precipitation (Bucheli-Witschel, 2001; Sutton, 1985; van der Maas et al., 2006). Additionally, Zhang et al. (2018) calculated the operating costs of a long-term two-stage CABR system and found that operating costs increased with increasing total iron concentration, and they observed that reducing the initial concentration of the chelating agent could inhibit EDTA iron loss.

In addition to the extra consumption generated from EDTA when the iron complex becomes unstable, Tucker et al. (1999) reported that the end products of the EDTA decomposition, such as ethylenediamine triacetic acid, iminodiacetic acid, and acetic acid, are released into the environment and generate pollution problems.

Meanwhile, the operation of CABR systems over long periods of time has been proven to be a result of the inhibition of microorganisms for the reduction of Fe(II)EDTA – NO^{2-} and Fe(III)EDTA$^-$, due to factors such as O_2 (Zhang et al., 2008a) and the accumulation of NO_2^-, NO_3^- (Zhou et al., 2013b), and sulfur compounds such as SO_3^{2-} (Manconi et al., 2006).

Iron complexes with NTA and citrate are less costly and toxic than those with EDTA; however, these compounds do not have the same absorption capacity as EDTA-based iron complexes, as reflected in the NO RE and $Fe(III)EDTA^-$ reduction. Recently, studies have focused on compounds containing cobalt, predominantly in terms of NO absorption. However, only a few batch experiments have been performed, and none have been performed at the laboratory scale (Sun et al., 2019a).

The use of alternative chelating agents or absorbents may be an option for the implementation of integrated CABR systems at an industrial scale. This new alternative should have an elevated NO absorption range, while being cost-efficient, renewable by microorganisms, and non-biodegradable. Moreover, it should not generate other compounds and should have low sensitivity to operational conditions.

5. Combination of CABR with other systems

5.1. Chemical absorption - bio electrochemical reduction (CABER)

5.1.1. System description

The use of a biofilm electrode reactor (BER) combined with chemical absorption by $Fe(II)EDTA^{2-}$, that is, chemical absorption-bio electrochemical reduction (CABER), was found to be an innovative alternative in the search for better $Fe(II)EDTA-NO^{2-}$ and $Fe(III)EDTA^-$ reduction (Guo et al., 2019).

Since the beginning of their usage in nitrate-polluted water treatments (Sakakibara et al., 1994; Sakakibara and Nakayama, 2001), BERs have further been proposed for the treatment of other pollutants, including NO. In a BER, the CO_2 generated in the anode (Eq. (8)), serves as a carbon source for bacterial growth and in the cathode (Eqs. (9) and (10)). Thus, microorganisms immobilized on the surface of the cathode use hydrogen gas as an electron acceptor generated by the electrolysis of water. The reactions that occur in the electrodes are as follows (Zhou et al., 2012):

Anodic reaction

$$C + 2H_2O_{(l)} \rightarrow CO_{2(g)} + 4H^+_{(aq)} + 4e^- \quad \text{(Eq. 8)}$$

Cathodic reactions

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH^-_{(aq)} \quad \text{(Eq. 9)}$$

$$^1/_2O_{2(g)} + 2e^- + H_2O_{(l)} \rightarrow 2OH^-_{(aq)} \quad \text{(Eq. 10)}$$

Furthermore, studies on $Fe(II)EDTA-NO^{2-}$ denitrification in BER systems can occur in two ways. The first is biologically, through an electron donor, in a process similar to that occurring in a conventional CABR system. The second method involves the use of $Fe(II)EDTA^{2-}$ as an electron donor (Eq. (11)) (Li et al., 2015a). In these systems, autotrophic denitrification is the main mechanism for $Fe(II)EDTA-NO^{2-}$ reduction (Zhao et al., 2018).

$$2Fe(II)EDTA-NO^{2-}_{(aq)} + Fe(II)EDTA^{2-}_{(aq)} + 4H^+_{(aq)} \xrightarrow{\text{microorganism}} 4Fe(III)EDTA^-_{(aq)} + N_{2(g)} + H_2O_{(l)} \quad \text{(Eq. 11)}$$

Li et al. (2015a) reported that $Fe(III)EDTA^-$ reduction in a BER can directly occur electrochemically (Eq. (12)), and indirectly via bioreduction using H_2 produced by H_2O electrolysis as an electron donor for reduction (Eq. (13)). This is one of the advantages of CABER systems because the addition of H_2 reduces electron donor consumption.

$$Fe(III)EDTA^-_{(aq)} + e^- \rightarrow Fe(II)EDTA^{2-}{}_{(aq)} \quad (e^0 = 96\ mV) \quad \text{(Eq. 12)}$$

$$2Fe(III)EDTA^-_{(aq)} + H_{2(aq)} + 2OH^-_{(aq)} \xrightarrow{\text{microorganism}} 2Fe(II)EDTA^{2-}_{(aq)} + 2H_2O_{(l)} \quad \text{(Eq. 13)}$$

Compared to CABR systems, CABER systems include a shorter gas residence time, higher NO elimination capacity, and greater tolerance for oxygen. Xia et al. (2014) conducted a comparative study of both systems and found that the CABER integrated system required an EBRT of only 20 s, whereas the EBRT required in the CABR integrated system for it to reach the same 90% RE NO was 45 s. As for NO elimination, the maximum EC in a CABER was 104.2 g NO m^{-3} h^{-1}, while in a CABR integrated system it was only 18.78 g NO m^{-3} h^{-1}. In the case of oxygen, CABER systems were not sensitive until the concentration of oxygen in the inlet gas was 10%.

Table 4 summarizes the existing experiences in CABER systems, where the types of systems, operating conditions, and maximum reductions of NO and $Fe(III)EDTA^-$ are presented.

New research on CABER systems is focused on exploring the use of a third electrode to improve $Fe(II)EDTA-NO^{2-}$ and $Fe(III)EDTA^-$ reduction efficiency. Zhou et al. (2012) compared two materials, activated carbon and graphite, using a three-dimensional (3D) BER. This study achieved a reduction of $Fe(II)EDTA-NO^{2-}$ with activated carbon and graphite levels of 92–94% and 90–93%, respectively. However, a $Fe(III)EDTA^-$ reduction was achieved for activated carbon in the range of 34–67% and of graphite in the range of 58–67%. Despite similar RE levels, graphite was determined to be the most suitable of the two materials because activated carbon can lead to fewer adherences of microorganisms, lower RE, and less stability.

As shown in Table 4, different types of inocula have been used in CABER or BER systems for the formation of biofilms in cathodes. Some of the inocula used were often mixed cultures of sludge from wastewater treatment plants and pure cultures, such as *Escherichia coli* sp. FR-2 (Xia et al., 2014; Zhao et al., 2018) and *Pseudomonas* sp. DN-2(Li et al., 2015a).

5.1.2. Drawbacks of CABER systems

One limitation observed in $Fe(II)EDTA-NO^{2-}$ reduction is that the hydrogen generated in the BER cathode (Eq. (8)) does not serve as the primary electron donor. Therefore, it is necessary to add an organic electron donor to enhance denitrification (Zhou et al., 2012). For example, Gao et al. (2011) reported an RE NO of 85% in a BER which operated continually with an electric current of 0.03 A, and with glucose as electron donor. The absence of electricity decreased the removal efficiency with an inlet of approximately 2.1 mM $Fe(II)EDTA-NO^{2-}$. Liu et al. (2021) found that the use of glucose as an electron donor is crucial

Table 4
Summary of studies with CABER systems.

System	Electron donor	Chelating agent form	Chelating agent concentration (mM)	T °C	pH	Impressed Current (A)	Volume current density A m^{-3}	Current Density mA cm^{-2}	Electrolyte rod	Maximum Removal Efficiency of NO (%)	Maximum Reduction of **Fe(III)EDTA$^-$** (%)	Inoculum	Reference
Batch	H^*	Fe(III)$EDTA^-$	3 5 15	45–55	6.8–7.2	0.12	n.a.	0.09–0.59	n.a.	n.a.	91	*Escherichia coli* strain FR-2	Mi et al. (2009)
Continous	Glucose H^*	Fe(II)$EDTA - NO^{2-}$ Fe(III)$EDTA^-$	2 ± 0.25	50	6.8	0.03	n.a.	n.a.	Graphite	85	78	*Escherichia coli* strain FR-2 and *Pseudomonas* sp. Strain DN-2	Gao et al. (2011)
Continous	Glucose H^*	Fe(II)$EDTA - NO^{2-}$ Fe(III)$EDTA^-$	4 8	50	6.8	0–0.04	30.53	n.a.	Graphite/ Activated carbon	94.2	67.4	*Escherichia coli* strain FR-2 and *Pseudomonas* sp. Strain DN-2	Zhou et al. (2012)
Continous	Glucose H^*	Fe(II)$EDTA^{2-}$	10	45–55	6.2–6.8	0–0.08	0–133.3	n.a.	Graphite	98	n.a.	*Escherichia coli* strain FR-2 and *Pseudomonas* sp. Strain DN-2	YXia et al. (2014)
Continous	Glucose H^*	Fe(III)$EDTA^-$	8	45–55	6.2–6.8	0.02–0.04	30.53	n.a.	Graphite	n.a.	76	*Escherichia coli* strain FR-2 and *Pseudomonas* sp. Strain DN-2	Li et al., 2015a
Continous	H^*	Fe(II)$EDTA^{2-}$	25	30	7	n.a.	n.a.	n.a.	n.a.	57.45 ± 2.2	61.7 ± 2.4	Sludge wastewater treatment plant	Sun et al. (2015)
Continous	Glucose H^*	Fe(III)$EDTA^-$ Fe(II)$EDTA - NO^{2-}$	10 2–3	50	6.8–7.0	0.04–0.08	n.a.	0.007 0.010 0.014	Graphite	n.a.	n.a.	Activated sludge of a sewage treatment plant enrichment	Zhao et al. (2018)
Continous	Glucose H^*	Fe(II)$EDTA^{2-}$ Fe(III)$EDTA^-$	30 100	50	6.8–7.2	0.02	n.a.	0.00009	Graphite/ Polypyrrole	n.a.	n.a.	*Escherichia coli* strain FR-2 and *Pseudomonas* sp. Strain DN-2	Guo et al. (2019)
Continous	Glucose H^*	Fe(II)$EDTA^{2-}$ Fe(III)$EDTA^-$	20	n.a	6.8	n.a.	22.9	n.a.	Graphite	98.35	98.35	Activated sludge of a sewage treatment plant enrichment	Liu et al. (2021)

n.a. not available.

for the regeneration of the chelating agent, while the current is only a promoting agent for this regeneration.

Furthermore, at high concentrations, $Fe(III)EDTA^-$ reduction can be affected by the microbial inhibition of $Fe(II)EDTA-NO^{2-}$, and vice versa (Sun et al., 2015; Zhou et al., 2012). In some reactors, when the operation was continuous and the reduction of $Fe(III)EDTA^-$ and $Fe(II)EDTA-NO^{2-}$ took place simultaneously, no high reduction efficiencies were obtained for either one or the other and were potentially achieved for neither. For example, Sun et al. (2015) achieved an RE of only 62% and 58% for $Fe(III)EDTA^-$ and $Fe(II)EDTA-NO^{2-}$, respectively.

Another significant drawback of CABER systems is the stability and self-renewal of the biofilm on the cathode surface, which is essential for NO removal and $Fe(III)EDTA^-$ regeneration (Gao et al., 2011; Zhao et al., 2018; Zhou et al., 2012). For example, up to 60 d have been reported for the immobilization of *Escherichia coli* sp. FR-2 (Mi et al., 2009) and it has taken up to 90 d to achieve a visible biofilm on the cathode surface (Gao et al., 2011).

5.2. Flue gas treatment with CABR: simultaneous abatement of SO_x and NO

5.2.1. Simultaneous abatement of SO_x and NO system description

Flue gases sometimes include sulfur compounds, mainly SO_2, which can be absorbed in aqueous phases, such as sulfite (SO_3^{2-}), and further oxidized to sulfate (SO_4^{2-}) (Eq. (14)). In biological flue gas desulfurization technologies (Bio-FGDs), SO_3^{2-} and SO_4^{2-} can be reduced to sulfide (S^{2-}) (Eq. (15)) by sulfate-reducing bacteria (SRB), which in the second stage are oxidized to elemental sulfur or sulfate by sulfide-oxidizing bacteria (SOB) (Pandey et al., 2005). The possibility of simultaneously reducing NO and SO_2 using desulfurization technology (Bio-FGD) and CABR systems has recently been studied.

$$SO_{2(g)} + H_2O_{(l)} \rightarrow H^+_{(aq)} + SO_3^{2-}(aq) \xrightarrow{O_2} SO_4^{2-}(aq) \quad \text{(Eq. 14)}$$

$$SO_3^{2-}(aq) \Big/ SO_4^{2-}(aq) \xrightarrow{SRB} S^{2-}_{(aq)} \xrightarrow{H^+} HS^-_{(aq)} \xrightarrow{H^+} H_2S_{(g)} \quad \text{(Eq. 15)}$$

In the simultaneous abatement of SO_x and NO_x, sulfide (produced in the desulfurization stage) can be used as an electron donor to enhance the ability of autotrophic denitrifying bacteria to reduce $Fe(II)EDTA-NO^{2-}$ (Eq. (17)) (Chen et al., 2019).

$$4Fe(II)EDTA-NO^{2-}_{(aq)} + HS^-_{(aq)} \xrightarrow{\text{microorganism}} 4Fe(II)EDTA^{2-}_{(aq)} + 2N_{2(g)} + SO_4^{2-}(aq) + H^+_{(aq)} \quad \text{(Eq. 16)}$$

One of the main advantages of this process is that it oxidizes sulfide to sulfate and thiosulfates to elemental sulfur and converts $Fe(II)EDTA-NO^{2-}$ into N_2. Sun et al. (2019a) showed that sulfide can be converted into elemental sulfur at different NO concentrations through oxidation–reduction reaction equations.

Furthermore, the elemental sulfur produced can be applied to agricultural and industrial sectors. For example, Zhang et al. (2019b) achieved 78% of elemental sulfur when the SO_4^{2-} concentration in the influent was 15 mM. The conversion of elemental sulfur from $Fe(II)EDTA-NO^{2-}$ and sulfide occurred via the sulfur autotrophic denitrification process, as follows:

$$Fe(II)EDTA-NO^{2-}_{(aq)} + HS^-_{(aq)} \xrightarrow{\text{microorganism}} Fe(II)EDTA^{2-}_{(aq)} + \frac{1}{2}N_{2(g)} + S^0_{(aq)} + OH^-{}_{(aq)} \quad \text{(Eq. 17)}$$

This process has only been studied at the laboratory-scale. Manconi et al. (2006) studied the simultaneous abatement of SO_x and NO through batch tests to assess the influence of SO_2 in various NO treatment systems. In continuous systems, the integration of SO_2 and NO removal has been studied in one-stage systems such as the BTF (van der Maas et al., 2006; Zhang et al., 2008a), RDB (Chen et al., 2016a, 2019), and moving-bed biofilm reactor (MBBR) (Zhang et al., 2019b). Two-stage systems have also been tested (Xu et al., 2022; Zhou et al., 2013b). A summary of the conditions tested for the simultaneous abatement of SO_x and NO is presented in Table 5.

Regarding the operating conditions of the reactors used in the simultaneous reduction of NO and SO_x, as shown in Table 5, some reactors were operated at temperatures ranging from 45 to 55 °C (Zhang et al., 2008a) and others at room temperature (Chen et al., 2013b); however, it has been reported that the solubility of SO_2 in water is higher at room temperature, allowing better RE to be achieved (Chen et al., 2016a). The optimal pH for $Fe(II)EDTA-NO^{2-}$ regeneration and S^0 recovery was found to be 8 (Xu et al., 2022).

In different studies, it was found that high concentrations of NO at the inlet positively affect the desulfurization process. Chen et al. (2016a) achieved an increase in SO_2 RE from 78% to 95%, with the inlet NO concentration increases up to 636 ppm_v in an RDB. This is because an increase in the inlet concentration leads to the formation of polysulfide, which can oxidize S^{2-} and benefit the reduction of dissolved SO_2 by SRB (Wang et al., 2013).

Regarding other operating parameters, Chen et al. (2016a) reported that O_2 increases potentially inhibit enzymatic activity in SRB bacteria and the oxidation of $Fe(II)EDTA^{2-}$, causing a decrease in NO and SO_2 RE. For this reason, these systems must be kept with oxygen concentrations below 2% v/v. Additionally, an appropriate EBRT was shown to be 108 s, which led to a maximum RE of SO_2 and NO of 99% and 93% for SO_2 and NO inlet concentrations of 763 ppm_v and 652 ppm_v, respectively.

Different microbial communities were found in the simultaneous abatement of the SO_x and NO systems. For example, Zhang et al. (2008a) found *Pseudomonas* sp. and *Escherichia coli* in a BTF that was inoculated with activated sludge from a municipal wastewater treatment plant, which led to a maximum removal capacity of 18.78 g NO m^{-3} h^{-1} during 5 h of operation at the steady state. Furthermore, Chen et al. (2016a, 2019) and Zhang et al. (2019b) showed that *Pseudomonas, Sulfurovum,* and *Arcobacter* were present in denitrification processes, whereas *Desulfovibrio* and *Desulfomicrobium* were present in sulfate reduction processes. Chen et al. (2015b) obtained a NO RE greater than 98% with

Table 5
Summary of studies testing simultaneous abatement of SO_2 and NO_x.

System/ reactor	Electron donor	Inlet (ppm_v)	Volumetric flow rate (m^3h^{-1})	T °C	pH	Presence of free oxygen O_2 (%)	EBRT (s)	Maximum Removal Efficiency of NO (%)	Elimination Capacity NO (Ec) ($g\ m^{-3}{\cdot}h^{-1}$)	Inoculum	Reference
Batch	Ethanol S^{2-}	n.a.	n.a.	37–55	6.8–7.4	Anaerobic	n.a.	n.a.	n.a.	Anaerobic methanogenic granular. denitrifying and BioDeNOx sludge - *Escherichia coli*	van der Maas et al. (2005b)
Batch	Ethanol S^{2-}	n.a.	n.a.	55	7.2	Anaerobic	n.a.	n.a.	n.a.	Anaerobic methanogenic granular sludge and denitrifying sludge from wasterwater treatment plants. Sludge of BioDeNOx reactor	Manconi et al. (2006)
Continuous/ BTF	Ethanol	NO: 60-155	1.5	55	7.0–7.4	3.5–3.9	13.2	40	5.35–13.81	Anaerobic methanogenic granular sludge and denitrifying sludge from wasterwater treatment	van der Maas et al. (2006)
Continuous/ BTF	Glucose	NO: 50-2959 SO_2: 76-305	0.06	45–55	n.a.	1–6.5	600	>70	0.14–18.78	Actived sludge wastewater treatment plant enrichment -*Pseudomonas sp.* and *Escherichia coli*	Zhang et al. (2008a)
Batch	Ethanol Methanol Acetate Glucose S^{2-}	n.a.	n.a.	55	7–7.4	Anaerobic	n.a.	n.a.	n.a.	Methanogenic granular sludge. denitrifying sludge and BioDeNOx sludge reactor	van der Mass et al. (2009)
Batch	Lactate	n.a.	n.a.	30	7	Anaerobic		90	n.a.	Sewage treatment plant - *Desulfovibrio* sp. *CMX*	Chen et al. (2013b)
Continuous/ two stages - sintered glass packed column + packed column	Glucose	NO:112–1062 SO_2: 927-966	0.06–0.12	40 ± 0.5	6.5–7.0	3–10	115–229	95	4.22–4.48	Actived sludge wastewater treatment plant enrichment	Zhou et al. (2013b)
Continuous/ two BTF	HS^-	NO: 2037-2445 SO_2:573-763	0.1	25–35	7.5	4–20	1620–1980	80.85	n.a	Activated sludge wastewater treatment plant enrichment	Wang et al. (2015)
Batch	Lactate	n.a.	n.a.	30	7	Anaerobic	n.a.	98	n.a.	*Desulfovibrio* sp. CMX	Chen et al. (2015b)
Continuous/ RDB	HS^-	NO: 147-978 SO_2: ~954	0.1–1.44	25–35	7	8	30–420	93	n.a.	Actived sludge wastewater treatment plant enrichment	Chen et al. (2016a)
Continuous/ RDB	HS^-	NO: 326–978 SO_2: 89 - 813	0.48	25–35	4.1–8.2	0–8	325	93	2.76–8.30	Actived sludge wastewater treatment plant enrichment	Chen et al. (2019)
Continuous/ MBBR	HS^-	n.a.	n.a.	30	7	Anaerobic	n.a.	92	n.a.	Sludge of a Sewage treatment plant	Zhang et al. (2019b)
Continuous/ BTF	Glucose	NO: 122-570 SO_2: 57-191	0.06	48 ± 2	7–7.4	3	144	98.08	12.25	Actived sludge wastewater treatment plant enrichment	Sun et al. (2019a)

(*continued on next page*)

Table 5 (*continued*)

System/ reactor	Electron donor	Inlet (ppm_v)	Volumetric flow rate (m^3h^{-1})	T °C	pH	Presence of free oxygen O_2 (%)	EBRT (s)	Maximum Removal Efficiency of NO (%)	Elimination Capacity NO (Ec) ($g\ m^{-3}\cdot h^{-1}$)	Inoculum	Reference
	$S_2O_3^{2-}$	n.a.	n.a.	22–23	7.4		n.a.	39	n.a	Denitrifying sludge	Wu et al. (2022)
Continuous/ SBR						Anoxic					
Continuous/ two stages – spray scrubber + EGSB	Glucose	NO: 500 SO_2: 2500	0.006–0.06	n.a.	6.9–7.1	1–3	450–4500	100	n.a.	Actived sludge wastewater treatment plant enrichment	Xu et al. (2022)

n.a. not available

enriched *Desulfovibrio* sp. CMX, in an anaerobic incubator containing bacteria. Xu et al. (2022) found symbiosis between denitrifying bacteria (*Saprospiraceae* and *Dechloromonas*) and iron-reducing bacteria (*Klebsiella* and *Petrimonas*) in an expanded granular sludge bed (EGSB) reactor.

5.2.2. Drawbacks of simultaneous abatement of SO_x and NO

One limiting factor in the simultaneous abatement of SO_x and NO is the accumulation of H_2S in the system, which can inhibit the process and produce odor emissions. Possible inhibition of the integrated process was observed by Zhou et al. (2013b) in a two-stage system in which 97% of SO_2 removal efficiency was achieved with a SO_2 concentration of >496 ppm_v; however, this percentage decreased with higher concentrations of SO_2, removing only 81% SO_2 with a concentration of 1832 ppm_v. This can be attributed to the increase in S^{2-} concentration in the liquid phase of the BIO-FGD-CABR system, which further inhibits denitrifying bacteria in the systems, leading to a decrease in NO removal efficiency (Zielke and Wu, 1986).

To minimize H_2S concentration, Chen et al. (2019) recommended that bioreactors must operate with high NO inlet concentrations, neutral or weakly alkaline pH conditions, and an upper O_2 concentration limit of approximately 2% v/v.

Shen et al. (2022) concluded in their review that simultaneous abatement of SOx and NO through biological processes has limitations due to high pressure drops and packing blockages in BTFs, instabilities in RDBs, low efficiencies in hydrophobic gases such as NO, and high construction costs in MBR.

6. Microalgae for NO abatement

The capture of CO_2 and NO as carbon and nitrogen sources, respectively, for biomass formation has also been exploited (Jin et al., 2008; Nagase et al., 2001). Previous studies have obtained promising results for NO removal, resulting in steady and reliable operation (Van Den Hende et al., 2012). Additionally, this alternative allows for the creation of valuable products from flue gases.

The NO capture pathway via microalgae has also been investigated. The first pathway occurs when NO is oxidized to nitrate in an aqueous medium by dissolved oxygen (Van Eynde et al., 2016). The dominant limiting factor of this pathway is the low NO solubility problem described in this review. The second pathway was described by Nagase et al. (2001), in which gaseous NO is directly taken up through diffusion into the microalgae cells.

6.1. Operational conditions of microalgae reactors

The use of microalgae for NO abatement has been studied mainly in long tubular photobioreactors, which have proven to be suitable for the joint removal of CO_2 and NO (Yoshihara et al., 1996). Other studies have been conducted on bubble columns and airlift reactors (Nagase et al., 1998).

Van Eynde et al. (2016) included a photocatalytic reactor before the photobioreactor to convert NO into NO_2, achieving an NO removal of 84% with a combustion gas of 1% CO_2 and 50 ppm_v of NO. To improve the removal of NO in the system, Nagase et al. (1997), rather than using high-dimension columns to increase the time and the gas-liquid contact area, reduced the size of the bubble, thereby increasing the contact area and leading to a higher NO reduction.

To improve the gas-liquid mass transfer and enhance NO fixation in microalgae, Jin et al. (2008) added an $Fe(II)EDTA^{2-}$ solution for the growth of *Scenedesmus* and reached only 40–45% RE NO since there was $Fe(III)EDTA^{-}$ oxidation, which cannot be regenerated.

An energy source is necessary to produce biomass from microalgae, which is normally light, because the absorption of NO depends on photosynthesis. Nagase et al. (1997) reported that cell growth is inhibited when cells are in contact with NO and not exposed to light.

Normally, the influence of temperature depends on the type of microalgae cultivated. For example, Ma et al. (2019) cultivated *Scenedesmus obliquus* sp. PF3, and found that it began to die at temperatures above 30 °C. Furthermore, they found that the optimum temperature in terms of maximum cell density for *Scenedesmus obliquus* sp. PF3 is 25 °C.

Another factor that plays an important role is pH. Flue gases containing NO can acidify the liquid phase, thereby inhibiting microalgae growth (Lee et al., 2000). A key inhibitor of microalgae growth is SO_2, which decreases the pH of the medium (Matsumoto et al., 1997). Additionally, López-Archilla et al. (2004) determined that the medium could be alkalized in the presence of NO_3^- and carbonates (HCO_3^-). Most microalgae have an optimal pH between 6.0 and 10 (Qie et al., 2019).

The main limiting factor in the application of this technique is the NO inlet concentration during the production of microalgae. One study using the marine microalgae strain NOA-113 achieved an NO removal efficiency of only 50% with a gas concentration of 300 ppm_v of NO and 15% v/v CO_2 in N_2 (Yoshihara et al., 1996). Additionally, *Scenedesmus obliquus* PF3 reached a maximum NO tolerance of 500 ppm_v in a photobioreactor (Ma et al., 2019). *Chlorella sorokiniana* was shown to be the microalgae with the highest NO tolerance capacity (577.5 ppm_v) and led to NO removal efficiencies approaching 95% (Lizzul et al., 2014; Qie et al., 2019).

Furthermore, studies have recommended that the gas flow rate must be reduced to obtain a high algae biomass fixation and an elevated RE (Du et al., 2019; Lee et al., 2000). This agrees with another study using the unicellular microalgae *Dunaliella tertiolecta*, which showed that gas flow rates between 100 and 400 mL/min and at a concentration of 100 ppm_v NO and 15% v/v CO_2 in gas N_2 led to an NO RE of 60%, which diminished when the inlet flow gas rate was increased (Nagase et al., 1997).

One of the biggest limitations in biomass production is the water source, as freshwater is generally used because it does not contain many pollutants that inhibit microalgae. Recently, Ma et al. (2020) explored

the possibility of using domestic wastewater with ammonium content for the biomass growth of the *Tetradesmus obliquus* PF3 culture, reaching an RE NO higher than 70%, and concluded that the use of this type of water may be feasible.

6.2. Microalgae involved

Several microalgae species have been used in studies where NO is a source of nitrogen. For example, Negoro et al. (1991) cultivated *Nannochloris* sp. NANNO2 and showed that biomass growth occurred at NO concentrations of up to 300 ppm_v. Nagase et al. (1998) captured NO from *Dunaliella tertiolecta* and achieved a maximum removal efficiency of 96% at a concentration of 100 ppm_v NO.

Various species of *Scenedesmus* have also been tested. Jiang et al. (2013) cultivated *Scenedesmus dimorphus*, leading to a CO_2 utilization efficiency of 76%, maximum biomass concentration of 3.63 g L^{-1}, and NO-concentration tolerance of 300 ppm_v. Li et al. (2015b) demonstrated that *Scenedesmus raciborskii* sp. WZKMT can be inhibited at a concentration of 200 ppm_v SO_2 and 100 ppm_v NO because it generates a decrease in pH. Ma et al. (2019) achieved a high NO RE of 97% by cultivating *Scenedesmus obliquus* sp. PF3.

Qie et al. (2019) revealed in their review that *Chlorella* has a higher commercial value owing to its high protein content; therefore, it is one of the most cultivated genera in NO capture research. For example, Kao et al. (2014) demonstrated that gases from different equipment in a steel plant can serve as a source of nitrogen and carbon for the cultivation of the microalgae *Chlorella* sp. MTF-15, having reached a NO RE of 95% in a hot stove and 80% in a power plant. Du et al. (2019) cultivated *Chlorella pyrenoidosa* sp. XQ-20044, leading to a NO RE of 84%, with a flue gas composition of 15% CO_2, 0.03% SO_2, and 0.03% NO. Other species tested included *Chlorella fusca* (Duarte et al., 2016).

6.3. Microalgae and circular economy

Scientific research on microalgae and flue gas treatment is related to the concept of a circular economy (Ma et al., 2014). This new approach, which seeks to find new and more sustainable forms of production and consumption, could lead to benefits, such as the reduction of negative environmental impacts, including CO_2 and NO emissions (Geng et al., 2019).

Depending on the type of microalgae cultivated for NO fixation, different functional components can be isolated from the biomass, such as fatty acids and lipids for the production of DHA/EPA biodiesel, sugars for the generation of bioethanol, butanol, and other chemical products of high value, proteins that can be used in the pharmaceutical industry, animal and aquaculture feed, and pigments that are used in natural foods (Yen et al., 2015).

Most studies have focused on biomass growth to produce biofuels because this could be more economically attractive than other applications. Kao et al. (2014) generated *Chlorella* sp. MTF-15 biomass, which has a high lipid composition and can be applied to biodiesel production. Additionally, Du et al. (2019) obtained the microalgal biomass of *Chlorella pyrenoidosa* sp. XQ-20044U with a total lipid content of 38% dry weight (DW) which is suitable as a biodiesel feedstock. Li et al. (2015b) cultivated *Scenedesmus raciborskii* sp. WZKMT with a high sugar content, which enables its use in bioethanol production. Lizzul et al. (2014) explored the possibility of coupling water treatment and microalgae production from the *Chlorella sorokiniana* culture to produce lipids, reaching n NO RE of 95–100%.

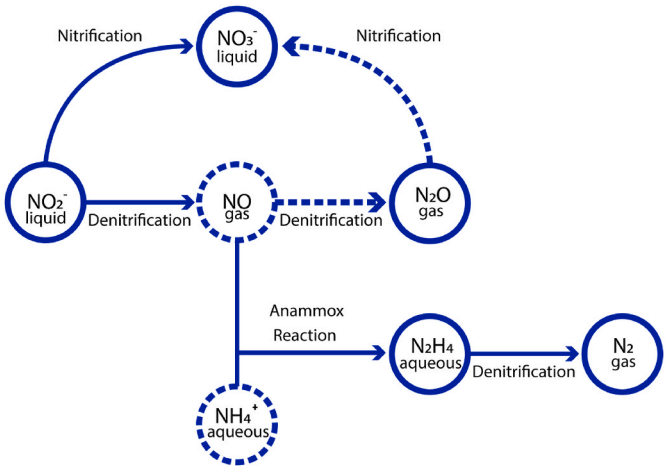

Fig. 4. Hypothetical anammox pathway with possible routes of NO removal (Adapted from Kartal et al., 2010).

Other functional compounds have also been explored; for example, Duarte et al. (2016) generated a biomass-cultivating species, *Chlorella fusca*, with a composition of 50% proteins that can be used by the pharmaceutical industry to produce biofilms, emulsifiers, and other products.

7. NO abatement through anammox bacteria

One process that is currently widely used for the removal of nitrogen in the wastewater treatment sector is anaerobic ammonium oxidation (anammox®). This technology consists of the biological oxidation of NH_4^+ to N_2 with NO_2^- as an electron acceptor performed by anaerobic ammonium oxidation bacteria (AnAOB) (Kartal et al., 2013). This process generates 100% savings in the organic carbon source and an extremely low sludge production compared to conventional denitrification processes (Henze et al., 2008). Additionally, it requires low energy and has highly efficient nitrogen removal ability (Wang et al., 2018).

Molecular studies of the anammox mechanism (Eq. (18) and (19)) have shown that NO can react with NH_4^+ to produce hydrazine (N_2H_4) via the hydrazine synthase (HZS) enzyme, and that N_2H_4 can be oxidized into nitrogen gas (N_2) by the hydrazine dehydrogenase (HDH) enzyme (Wang et al., 2018). Furthermore, anammox bacteria are not inhibited by high concentrations of NO (Schmidt et al., 2002). Kartal et al. (2010) studied the effects of NO on anammox bacteria and found that the use of NO as an electron facilitated NH_4^+ removal in large-scale anammox bioreactors (Fig. 4).

$$NO_{(g)} + NH_4^+(aq) \rightarrow 0.25N_2H_{4(aq)} + H_2O_{(l)} + 0.75N_{2(g)} + H^+_{(aq)} \quad \text{(Eq. 18)}$$

$$N_2H_{4(aq)} \rightarrow N_{2(g)} + 4H^+_{(aq)} + 4e^- \quad \text{(Eq. 19)}$$

The integration of the annamox process and NO removal also requires the use of an absorbent or chelating agent to improve the NO mass transfer from the gas to liquid phases. Throughout all studies concerning anammox-NO removal integration, only $Fe(II)EDTA^{2-}$ has been studied. When $Fe(II)EDTA^{2-}$ is used, another reaction mechanism may coexist, as follows (Zhang et al., 2017):

$$6Fe(II)EDTA - NO^{2-}_{(aq)} + 4NH_4^+(aq) \xrightarrow{\text{AnAOB}} 6Fe(II)EDTA^{2-}_{(aq)} + 6H_2O_{(l)} + 5N_{2(g)} + 4H^+_{(aq)} \quad \text{(Eq. 20)}$$

7.1. Reactor operation of NO abatement through anammox bacteria using $Fe(II)EDTA^{2-}$

NO abatement through anammox bacteria treatment has been studied predominantly through different one-stage configurations, such as the use of a sequencing batch reactor (SBR) (Zhang et al., 2017) and an up-flow bioreactor with a sponge iron bed (Zhang et al., 2019a). Some two-stage configurations have also been tested using an absorption column and different reactors, including an up-flow anaerobic sludge blanket (UASB) reactor (Wang et al., 2016) and a biofilter with a polyhedron empty ball carrier (Wang et al., 2018). Studies have recommended that these reactors operate at a temperature of 25–45 °C and a pH between 6.5 and 8.0 (Zhang et al., 2017).

Wang et al. (2016) determined the appropriate concentration range of the $Fe(II)EDTA^{2-}$ complex to be between 0.0 and 3.5 mM for anammox bacteria to maintain an optimal NH_4^+ removal through $Fe(II)EDTA - NO^{2-}$ reduction. Wang et al. (2018) concluded that these systems require low concentrations of $Fe(II)EDTA^{2-}$ and do not require the addition of organic matter or dissolved oxygen to maintain the function and activity of anammox bacteria.

Wang et al. (2018) studied the influence of the velocity of the inlet liquid and found that when the velocity was increased to 79 mL/min, a maximum RE NO of 80% was reached. Additionally, Wang et al. (2016) reported that a complex-circulating time of 60 min is required to maximize the RE.

Zhang et al. (2017) reported that the parameters that most affected the long-term stability of anammox reactors were NO, Fe, and EDTA toxicity; therefore, it is necessary to add more anammox biomass to bioreactors. A maximum RE NO of 87% was reached compared to when no additional biomass was added, which decreased the RE NO to 44%.

One limitation of these systems is, as in CABR systems, the regeneration of $Fe(II)EDTA^{2-}$. Therefore, Zhang et al. (2019a coupled a spongy iron bed to an autotrophic upflow bioreactor and achieved an average Fe^{2+} loss rate of 0.190 mM/d and an average NO removal rate of 7.57 mM/d. Although it is a promising technology, there are only a few studies in the literature in which different operating parameters and their feasibility for scaling have been studied.

7.2. Microorganisms involved in NO abatement through anammox bacteria

Various researchers have analyzed microbial communities to determine bacterial diversity in the reactors used. Most have identified that anammox bacteria, namely *Candidatus Kuenenia*, *Candidatus Brocadia*, and *Candidatus Jettenia*, survive in an environment with combustion gases containing NO. Additionally, they concluded that the increasing abundance of these microorganisms is related to NO reduction through the anammox process (Wang et al., 2016, 2018; Zhang et al., 2017).

In addition to anammox bacteria, other communities such as denitrifying bacteria have been found. For example, Zhang et al., (2019a found that *Thauera*, *Hyphomicrobium*, *Azoarcus*, and *Azospir* are typical denitrifying heterotrophic bacteria. Furthermore, researchers have found that some bacteria consume ammonium to metabolize NO_2^-. For example, Wang et al. (2018) found that species of nitrosobacteria (*Nitrosomonas* and *Nitrospira*) are capable of growing mixotropically on ammonia and hydroxylamine.

The formation of a symbiotic system of anammox and denitrifying bacteria, among others, has been shown to facilitate NO removal. Wang et al. (2018) found that symbiosis of the system between denitrifying bacteria and anammox can alleviate the effects of inhibitors such as oxygen and organic matter. Furthermore, Zhang et al., (2019a reported that it is beneficial to have denitrifying bacteria in a system with anammox bacteria that can reduce $Fe(III)EDTA^-$ and thus increase the efficiency of the system.

8. Conclusions

Conventional techniques for NO abatement, such as nitrification, have not yet been widely explored; however, advances have been made in denitrification processes since the implementation of MBRs, which improve the solubility of NO and lead to a greater RE NO.

The use of $Fe(II)EDTA^{2-}$ as a chelating agent in CABR systems can be oxidized and highly unstable. Although operating parameters have been widely studied and optimized at the laboratory scale, there is no evidence that they can be scaled at an industrial level. Other complexes, such as NTA or citrate, have been studied, but they present disadvantages in terms of RE NO to be able to replace $Fe(II)EDTA^{2-}$.

The combination of CABR systems with other electrochemical systems such as CABER has also been explored. Despite the advantage that fewer electron donors must be used, their efficiencies are still very low.

Although the simultaneous abatement of SO_x and NO has not been widely studied and has limitations due to the accumulation of H_2S in the system, it is an attractive technology because the two pollutants can be treated simultaneously. Moreover, it has added value, namely, the production of elemental sulfur.

NO capturing through microalgae is one of the technologies currently of interest to researchers; however, its main disadvantages are its sensitivity to high concentrations of NO, the cost of energy and photobioreactors, and the necessity of a mass transfer vector to increase fixation. The biomass generated has different applications, which are attractive to the food, pharmaceutical, and energy industries, in line with the concept of a circular economy.

It is also a promising technique for the integration of the NO reduction process with other wastewater elimination processes, such as anammox reactors, thus reducing costs and taking advantage of other waste; however, it is evident in the literature that there is a lack of studies on different operational parameters such as residence time, NO inlet concentration, and inhibition of anammox biomass, meaning that this technique has low technology readiness levels (TRL).

Biological technologies for NO_x abatement are promising. Nevertheless, resources must be included in their demonstration (optimization) at the pilot scale in a real environment. Additionally, costs must be accurately calculated because their implementation will only be viable if they are economically compatible with current (physicochemical) systems.

Future research should focus on different aspects for these techniques to be implemented at pilot and industrial scales. It is essential to enhance the mass transfer of NO from the gas to the liquid phase. This can be accomplished in two ways. First, a mass transfer vector that can be converted must be found so that this compound can be recovered in a physicochemical or biological manner, without generating high costs in the process. The second method is in the design and operation of bioreactors, where it must be established whether a one-stage or two-stage configuration generates better elimination capacity and removal efficiency of NO.

Author contribution

Conceptualization: David Cubides, Xavier Guimerà, Irene Jubany and Xavier Gamisans. **Methodology**: David Cubides. **Validation**: Xavier Guimerà, Irene Jubany and Xavier Gamisans. **Formal analysis:** David Cubides and Xavier Guimerà **Investigation:** David Cubides. **Data curation:** David Cubides. **Writing—original draft preparation:** David Cubides and Xavier Guimerà. **Writing—review and editing:** Irene Jubany and Xavier Gamisans. **Visualization:** David Cubides. **Supervision:** Xavier Guimerà, Irene Jubany and Xavier Gamisans. **Project administration:** Irene Jubany and Xavier Gamisans. Funding **Acquisition:** Irene Jubany and Xavier Gamisans. All authors have read and agreed to the published version of the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

Data availability

No data was used for the research described in the article.

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References

Abdulrasheed, A.A., Jalil, A.A., Triwahyono, S., Zaini, M.A.A., Gambo, Y., Ibrahim, M., 2018. Surface modification of activated carbon for adsorption of SO_2 and NOX : a review of existing and emerging technologies. Renew. Sustain. Energy Rev. 94, 1067–1085. https://doi.org/10.1016/j.rser.2018.07.011.

Adewuyi, Y.G., Sakyi, N.Y., 2013. Simultaneous absorption and oxidation of nitric oxide and sulfur dioxide by aqueous solutions of sodium persulfate activated by temperature. Ind. Eng. Chem. Res. 52, 11702–11711. https://doi.org/10.1021/ie401649s.

Anthony, J.L., Anderson, J.L., Maginn, E.J., Brennecke, J.F., 2005. Anion effects on gas solubility in ionic liquids. J. Phys. Chem. B 109, 6366–6374. https://doi.org/10.1021/jp046404l.

Apel, W.A., Turick, C.E., 1993. The use of denitrifying bacteria for the removal of nitrogen oxides from combustion gases. Fuel 72, 1715–1718. https://doi.org/10.1016/0016-2361(93)90360-E.

Arnold, R.G., Hoffmann, M.R., DiChristina, T.J., Picardal, F.W., 1990. Regulation of dissimilatory Fe(III) reduction activity in *Shewanella putrefaciens*. Appl. Environ. Microbiol. 56, 2811–2817. https://doi.org/10.1128/aem.56.9.2811-2817.1990.

Barnes, J.M., Apel, W.A., Barrett, K.B., 1995. Removal of nitrogen oxides from gas streams using biofiltration. J. Hazard Mater. 41, 315–326. https://doi.org/10.1016/0304-3894(94)00103-N.

Baukal, C., 2005. Everything you need to know about NOx. Met. Finish. 103, 18–24. https://doi.org/10.1016/S0026-0576(05)80816-5.

Baukal, C., 2004. Industrial Combustion Pollution and Control. M. Dekker, New York.

Begel, S., Puchta, R., Sutter, J., Heinemann, F.W., Dahlenburg, L., Eldik, R. Van, 2015. Studies on the reaction of iron(II) with NO in a noncoordinating ionic liquid. Inorg. Chem. 54, 6763–6775. https://doi.org/10.1021/acs.inorgchem.5b00595.

Bouwman, A.F., Beusen, A.H.W., Griffioen, J., Van Groenigen, J.W., Hefting, M.M., Oenema, O., Van Puijenbroek, P.J.T.M., Seitzinger, S., Slomp, C.P., Stehfest, E., 2013. Global trends and uncertainties in terrestrial denitrification and N_2O emissions. Philos. Trans. R. Soc. B Biol. Sci. 368, 1–11. https://doi.org/10.1098/rstb.2013.0112.

Bucheli-Witschel, M., 2001. Environmental fate and microbial degradation of aminopolycarboxylic acids. FEMS Microbiol. Rev. 25, 69–106. https://doi.org/10.1016/S0168-6445(00)00055-3.

Buisman, C.J.N., Dijkman, H., Verbraak, P.L., Den Hartog, A.J., 2001. Apparatus for Purifying Flue Gas Containing Nitrogen Oxides. U.S.

Cai, W.-J., Tang, Z.-P., Li, J.-W., 2015. Removal of nitric oxide from simulated gas by the corona discharge combined with cobalt ethylenediamine solution. Fuel Process. Technol. 140, 82–87. https://doi.org/10.1016/j.fuproc.2015.08.035.

Cao, N., Gan, L., Xiao, Q., Lv, X., Lin, W., Li, H., Wang, C., 2020. Highly efficient and reversible nitric oxide capture by functionalized ionic liquids through multiple-site absorption. ACS Sustain. Chem. Eng. 8, 2990–2995. https://doi.org/10.1021/acssuschemeng.9b07657.

Carriazo, D., Serrano, M.C., Gutiérrez, M.C., Ferrer, M.L., del Monte, F., 2012. Deep-eutectic solvents playing multiple roles in the synthesis of polymers and related materials. Chem. Soc. Rev. 41, 4996–5014. https://doi.org/10.1039/C2CS15353J.

Chagnot, E., Taha, S., Martin, G., Vicard, J.F., 1998. Treatment of nitrogen oxides on a percolating biofilter after pre- concentration on activated carbon. Process Biochem. 33, 617–624. https://doi.org/10.1016/S0032-9592(98)00024-7.

Chandrashekhar, B., Pai, P., Morone, A., Sahu, N., Pandey, R.A., 2013. Reduction of NOx in Fe-EDTA and Fe-NTA solutions by an enriched bacterial population. Bioresour. Technol. 130, 644–651. https://doi.org/10.1016/j.biortech.2012.12.051.

Chandrashekhar, B., Pandey, R.A., 2017. Experimental and modelling study of treatment and regeneration of ferrous-nitrilotriacetate solution scrubbed with nitric oxide by an up-flow anaerobic biofilm reactor. J. Clean. Prod. 155, 179–188. https://doi.org/10.1016/j.jclepro.2016.09.013.

Chandrashekhar, B., Sahu, N., Tabassum, H., Pai, P., Morone, A., Pandey, R.A., 2015. Treatment of ferrous-NTA-based NOx scrubber solution by an up-flow anaerobic packed bed bioreactor. Appl. Microbiol. Biotechnol. 99, 5281–5293. https://doi.org/10.1007/s00253-014-6372-5.

Chen, H., Yang, C., Zeng, G., Yu, K., Qu, W., Yu, G., Meng, L., 2007. Numerical simulation for volatile organic compound removal in rotating drum biofilter. Chin. Sci. Bull. 52, 2184–2189. https://doi.org/10.1007/s11434-007-0332-8.

Chen, J., Ma, J.F., 2006. Abiotic and biological mechanisms of nitric oxide removal from waste air in biotrickling filters. J. Air Waste Manag. Assoc. 56, 32–36. https://doi.org/10.1080/10473289.2006.10464433.

Chen, J., Wang, J., Ma, J., 2006a. Effects of gas flow-rate and inlet concentration on nitric oxide removal in an autotrophic biofilter. J. Chem. Technol. Biotechnol. 81, 812–816. https://doi.org/10.1002/jctb.1463.

Chen, J., Wang, Z.Y., Jiang, Y.F., Qian, H.F., Zhang, W., Chen, J.M., 2009a. Assessment of the bacterial community for denitrifying removal of nitric oxide in a rotating drum biofilter by denaturing gradient gel electrophoresis. Environ. Eng. Sci. 26, 1189–1196. https://doi.org/10.1089/ees.2008.0153.

Chen, J., Wu, C., Wang, J., Ma, J., 2006b. Performance evaluation of biofilters packed with carbon foam and lava for nitric oxide removal. J. Hazard Mater. 137, 172–177. https://doi.org/10.1016/j.jhazmat.2006.01.049.

Chen, J., Dai, Q.Z., Qian, H.F., Jiang, Y.F., Chen, Jianmeng, 2013a. Nitric oxide enhanced reduction in a rotating drum biofilter coupled with absorption by FeII (EDTA). J. Chem. Technol. Biotechnol. 88, 579–584. https://doi.org/10.1002/jctb.3865.

Chen, Jun, Gu, S., Zheng, J., Chen, Jianmeng, 2016a. Simultaneous removal of SO_2 and NO in a rotating drum biofilter coupled with complexing absorption by FeII(EDTA). Biochem. Eng. J. 114, 87–93. https://doi.org/10.1016/j.bej.2016.06.027.

Chen, Jun, Jiang, Y., Chen, Jianmeng, Sha, H., Zhang, W., 2009b. Dynamic model for nitric oxide removal by a rotating drum biofilter. J. Hazard Mater. 168, 1047–1052. https://doi.org/10.1016/j.jhazmat.2009.02.159.

Chen, Jun, Li, B., Zheng, J., Chen, Jianmeng, 2019. Control of H_2S generation in simultaneous removal of NO and SO_2 by rotating drum biofilter coupled with FeII (EDTA). Environ. Technol. 40, 1576–1584. https://doi.org/10.1080/09593330.2018.1426640.

Chen, J., Li, Y., Hao, H. hong, Zheng, J., Chen, Jian meng, 2015a. Fe(II)EDTA-NO reduction by a newly isolated thermophilic Anoxybacillus sp. HA from a rotating drum biofilter for NOx removal. J. Microbiol. Methods 109, 129–133. https://doi.org/10.1016/j.mimet.2014.12.015.

Chen, Jun, Wang, J., Zheng, J., Chen, Jianmeng, 2016b. Prediction and inhibition of the N_2O accumulation in the BioDeNOx process for NOx removal from flue gas. Bioproc. Biosyst. Eng. 39, 1859–1865. https://doi.org/10.1007/s00449-016-1660-3.

Chen, Jun, Wu, J., Wang, J., Zhang, S., Chen, Jianmeng, 2018. A mass-transfer model of nitric oxide removal in a rotating drum biofilter coupled with FeII(EDTA) absorption. Ind. Eng. Chem. Res. 57, 8144–8151. https://doi.org/10.1021/acs.iecr.8b00966.

Chen, M., Zhang, Y., Zhou, J., Dong, X., Wang, X., Shi, Z., 2013b. Sulfate removal by *Desulfovibrio* sp. CMX in chelate scrubbing solutions for NO removal. Bioresour. Technol. 143, 455–460. https://doi.org/10.1016/j.biortech.2013.06.037.

Chen, M., Zhou, J., Zhang, Y., Wang, Xiaojun, Shi, Z., Wang, Xiaowei, 2015b. Fe(III) EDTA and Fe(II)EDTA-NO reduction by a sulfate reducing bacterium in NO and SO_2 scrubbing liquor. World J. Microbiol. Biotechnol. 31, 527–534. https://doi.org/10.1007/s11274-015-1813-6.

Ciais, P., Sabine, C., Bala, G., Bopp, L., Brovkin, V., Canadell, J., Chhabra, A., DeFries, R., Galloway, J., Heimann, M., Jones, C., Quéré, C. Le, Myneni, R.B., Piao, S., Thornton, P., 2013. Carbon and other biogeochemical cycles. In: Climate Change 2013, pp. 465–570. https://doi.org/10.1017/CBO9781107415324.015.

Demmink, J.F., 2000. Removal of Hydrogen Sulfide and Nitric Oxide with Iron Chelates. University of Groningen.

Demmink, J.F., Van Gils, I.C.F., Beenackers, A.A.C.M., 1997. Absorption of nitric oxide into aqueous solutions of ferrous chelates accompanied by instantaneous reaction. Ind. Eng. Chem. Res. 36, 4914–4927. https://doi.org/10.1021/ie9702800.

Dilmore, R., Neufeld, R.D., Hammack, R.W., 2007. Kinetics of chemoheterotrophic microbially mediated reduction of ferric EDTA and the nitrosyl adduct of ferrous EDTA for the treatment and regeneration of spent nitric oxide scrubber liquor. Water Environ. Res. 79, 479–487. https://doi.org/10.2175/106143006x115390.

Dilmore, R., Neufeld, R.D., Hammack, R.W., 2006. Laboratory-scale iron EDTA-based NOX scrubbing process with biological treatment and regeneration of spent scrubber water. Environ. Eng. Sci. 23, 788–802. https://doi.org/10.1089/ees.2006.23.788.

Dong, X., Zhang, Y., Zhou, J., Chen, M., Wang, X., Shi, Z., 2013. Fe(II)EDTA-NO reduction coupled with Fe(II)EDTA oxidation by a nitrate- and Fe(III)-reducing bacterium. Bioresour. Technol. 138, 339–344. https://doi.org/10.1016/j.biortech.2013.03.181.

Dong, X., Zhang, Y., Zhou, J., Li, H., Wang, X., Chen, M., 2014. Evaluation of simultaneous reduction of Fe(II)EDTA-NO and Fe(III)EDTA by a bacterial pure culture. J. Chem. Technol. Biotechnol. 89, 111–116. https://doi.org/10.1002/jctb.4112.

Dong, X., Zhang, Y., Zhou, J., Li, N., Chen, M., 2012. Reduction of Fe(III)EDTA in a NOx scrubber liquor by a denitrifying bacterium and the effects of inorganic sulfur compounds on this process. Bioresour. Technol. 120, 127–132. https://doi.org/10.1016/j.biortech.2012.06.005.

Du, K., Wen, X., Wang, Z., Liang, F., Luo, L., Peng, X., Xu, Y., Geng, Y., Li, Y., 2019. Integrated lipid production, CO_2 fixation, and removal of SO_2 and NO from simulated flue gas by oleaginous *Chlorella pyrenoidosa*. Environ. Sci. Pollut. Res. 26, 16195–16209. https://doi.org/10.1007/s11356-019-04983-9.

Du Plessis, C.A., Kinney, K.A., Schroeder, E.D., Chang, D.P.Y., Scow, K.M., 1998. Denitrification and nitric oxide reduction in an aerobic toluene- treating biofilter.

Biotechnol. Bioeng. 58, 408–415. https://doi.org/10.1002/(SICI)1097-0290(19980520)58:4<408. AID-BIT8>3.0.CO;2-N.
Duarte, J.H., Fanka, L.S., Costa, J.A.V., 2016. Utilization of simulated flue gas containing CO_2, SO_2, NO and ash for Chlorella fusca cultivation. Bioresour. Technol. 214, 159–165. https://doi.org/10.1016/j.biortech.2016.04.078.
Durmazpinar, S., Ilhan, N., Demir, G., Insel, G., Dizge, N., Ergenekon, P., Erhan, E., Keskinler, B., 2014. Biological NOx removal by denitrification process in a jet-loop bioreactor: system performance and model development. Environ. Technol. 35, 1358–1366. https://doi.org/10.1080/09593330.2013.868529.
Ergas, S.J., 2001. Membran bioreactors. In: Bioreactors for Waste Gas Treatment. Kluwer Academic, pp. 163–164.
European Commission, 2013. Best Available Techniques (BAT) Reference Document for Waste Incineration, SpringerBriefs in Applied Sciences and Technology. https://doi.org/10.1007/978-1-4614-5752-7_3.
European Environment Agency, E., 2018. National Emission Ceilings Directive.
European Environment Agency, E., 2017. Air Quality in Europe — 2017, EEA Technical Report. https://doi.org/10.2800/22775.
Flanagan, W.P., Apel, W.A., Barnes, J.M., Lee, B.D., 2002. Development of gas phase bioreactors for the removal of nitrogen oxides from synthetic flue gas streams. Fuel 81, 1953–1961. https://doi.org/10.1016/S0016-2361(02)00130-8.
Frare, L.M., Vieira, M.G.A., Silva, M.G.C., Pereira, N.C., Gimenes, M.L., 2010. Hydrogen sulfide removal from biogas using Fe/EDTA solution: gas/liquid contacting and sulfur formation. Environ. Prog. Sustain. Energy 29, 34–41. https://doi.org/10.1002/ep.10374.
Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S. P., Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A., Karl, D.M., Michaels, A. F., Porter, J.H., Townsend, A.R., Vörösmarty, C.J., 2004. Nitrogen cycles: past, present and future. Biogeochemistry 70, 153–226.
Gambardella, F., Alberts, M.S., Winkelma, J.G.M., Heeres, E.J., 2005. Experimental and modeling studies on the absorption of NO in aqueous ferrous EDTA solutions. Ind. Eng. Chem. Res. 44, 4234–4242. https://doi.org/10.1021/ie048767d.
Gambardella, F., Winkelman, J.G.M., Heeres, H.J., 2006. Experimental and modelling studies on the simultaneous absorption of NO and O_2 in aqueous iron chelate solutions. Chem. Eng. Sci. 61, 6880–6891. https://doi.org/10.1016/j.ces.2006.07.003.
Gao, L., Mi, X.H., Zhou, Y., Li, W., 2011. A pilot study on the regeneration of ferrous chelate complex in NOx scrubber solution by a biofilm electrode reactor. Bioresour. Technol. 102, 2605–2609. https://doi.org/10.1016/j.biortech.2010.10.012.
García, G., Aparicio, S., Ullah, R., Atilhan, M., 2015. Deep eutectic solvents: physicochemical properties and gas separation applications. Energy Fuel. 29, 2616–2644. https://doi.org/10.1021/ef5028873.
Geng, Y., Sarkis, J., Bleischwitz, R., 2019. How to globalize the circular economy. Nature 565, 153–155. https://doi.org/10.1038/d41586-019-00017-z.
Guo, T., Zhang, C., Zhao, J., Ma, C., Li, S., Li, W., 2019. Evaluation of polypyrrole-modified bioelectrodes in a chemical absorption-bioelectrochemical reduction integrated system for NO removal. Sci. Rep. 9, 1–11. https://doi.org/10.1038/s41598-019-49610-2.
Han, L., Shaobin, H., Zhendong, W., Pengfei, C., Yongqing, Z., 2016. Performance of a new suspended filler biofilter for removal of nitrogen oxides under thermophilic conditions and microbial community analysis. Sci. Total Environ. 562, 533–541. https://doi.org/10.1016/j.scitotenv.2016.04.084.
Henze, M., van Loosdrecht, M., Ekama, G., Brdjanovic, D., 2008. Biological Wastewater Treatment: Principles, Modeling and Design. https://doi.org/10.2166/9781780408613.
Hishinuma, Y., Kaji, R., Akimoto, H., Nakajima, F., Mori, T., Kamo, T., Arikawa, Y., Nozawa, S., 1979. Reversible binding of NO to FE(II)EDTA. Bull. Chem. Soc. Jpn. 52, 2863–2865. https://doi.org/10.1246/bcsj.52.2863.
Hollocher, T.C., 1983. The pathway of nitrogen and reductive enzymes of denitrification. Antonie Leeuwenhoek 48, 531–544. https://doi.org/10.1007/BF00399539.
Huasheng, L., Wenchi, F., 1988. Kinetics of absorption of nitric oxide in aqueous iron(II)-EDTA solution. Ind. Eng. Chem. Res. 27, 770–774. https://doi.org/10.1021/ie00077a009.
Itahashi, S., Yumimoto, K., Kurokawa, J.I., Morino, Y., Nagashima, T., Miyazaki, K., Maki, T., Ohara, T., 2019. Inverse estimation of NOx emissions over China and India 2005-2016: contrasting recent trends and future perspectives. Environ. Res. Lett. 14 https://doi.org/10.1088/1748-9326/ab4d7f.
Jeter, R.M., Ingraham, J.L., 1981. In: Starr, M.P., Stolp, H., Trüper, H.G., Balows, A., Schlegel, H.G. (Eds.), The Denitrifying Prokaryotes BT - the Prokaryotes: A Handbook on Habitats, Isolation, and Identification of Bacteria. Springer Berlin Heidelberg, Berlin, Heidelberg, pp. 913–925. https://doi.org/10.1007/978-3-662-13187-9_73.
Jezwska-Trzebiatowska, B., Gerega, K., Formicka-Kozłowska, G., 1980. Effect of side amino acid groups on the effectiveness of nitric oxide binding by the cobalt complexes with amino acids. Inorg. Chim. Acta. 40, 187–190. https://doi.org/10.1016/S0020-1693(00)92002-4.
Jiang, R., Huang, S., Yang, J., Deng, K., Liu, Z., 2009. Field applications of a bio-trickling filter for the removal of nitrogen oxides from flue gas. Biotechnol. Lett. 31, 967–973. https://doi.org/10.1007/s10529-009-9969-5.
Jiang, W., Xu, Q., Wei, X., 2019. Use of cobalt(II) chelates of monothiol-containing ligands for the removal of nitric oxide. J. Hazard Mater. 374, 50–57. https://doi.org/10.1016/j.jhazmat.2019.04.025.
Jiang, Y., Zhang, W., Wang, J., Chen, Y., Shen, S., Liu, T., 2013. Utilization of simulated flue gas for cultivation of *Scenedesmus dimorphus*. Bioresour. Technol. 128, 359–364. https://doi.org/10.1016/j.biortech.2012.10.119.
Jin, H.F., Santiago, D.E.O., Park, J., Lee, K., 2008. Enhancement of nitric oxide solubility using Fe(II)EDTA and its removal by green algae *Scenedesmus* Sp. Biotechnol. Bioproc. Eng. 13, 48–52. https://doi.org/10.1007/s12257-007-0164-z.
Jin, Y., Veiga, M.C., Kennes, C., 2005. Bioprocesses for the removal of nitrogen oxides from polluted air. J. Chem. Technol. Biotechnol. 80, 483–494. https://doi.org/10.1002/jctb.1260.
Jinchao, W., Ping, Y., Bo, C., Yunbai, L., Houzhang, T., 2009. Removal of dilute nitric oxide using cobalt diethylenetriamine solution under aerobic condition. Separ. Sci. Technol. 44, 1590–1603. https://doi.org/10.1080/01496390902775349.
Jing, G., Zhou, J., Zhou, Z., Lin, T., 2012. Reduction of Fe(III)EDTA$^-$ in a NOx scrubbing solution by magnetic Fe_3O_4-chitosan microspheres immobilized mixed culture of iron-reducing bacteria. Bioresour. Technol. 108, 169–175. https://doi.org/10.1016/j.biortech.2011.12.112.
Jing, G.H., Li, W., Shi, Y., Ma, B.Y., Tan, T.E., 2004. Regeneration of nitric oxide chelate absorption solution by two heterotrophic bacterial strains. J. Zhejiang Univ. - Sci. 5, 432–435. https://doi.org/10.1631/jzus.2004.0432.
Johnston, H.L., Giauque, W.F., 1929. The heat capacity of nitric oxide from 14°k. to the boiling point and the heat of vaporization. vapor pressures of solid and liquid phases. the entropy from spectroscopic data. J. Am. Chem. Soc. 51, 3194–3214. https://doi.org/10.1021/ja01386a004.
Kampf, N., 2002. The use of polymers for coating of cells. Polym. Adv. Technol. 13, 895–904. https://doi.org/10.1002/pat.277.
Kantarci, N., Borak, F., Ulgen, K.O., 2005. Bubble column reactors. Process Biochem. 40, 2263–2283. https://doi.org/10.1016/j.procbio.2004.10.004.
Kao, C.Y., Chen, T.Y., Chang, Y. Bin, Chiu, T.W., Lin, H.Y., Chen, C. Da, Chang, J.S., Lin, C.S., 2014. Utilization of carbon dioxide in industrial flue gases for the cultivation of microalga *Chlorella* sp. Bioresour. Technol. 166, 485–493. https://doi.org/10.1016/j.biortech.2014.05.094.
Kartal, B., De Almeida, N.M., Maalcke, W.J., Op den Camp, H.J.M., Jetten, M.S.M., Keltjens, J.T., 2013. How to make a living from anaerobic ammonium oxidation. FEMS Microbiol. Rev. 37, 428–461. https://doi.org/10.1111/1574-6976.12014.
Kartal, B., Tan, N.C.G., Van de Biezen, E., Kampschreur, M.J., Van Loosdrecht, M.C.M., Jetten, M.S.M., 2010. Effect of nitric oxide on anammox bacteria. Appl. Environ. Microbiol. 76, 6304–6306. https://doi.org/10.1128/AEM.00991-10.
Kennes, C., Rene, E.R., Veiga, M.C., 2009. Bioprocesses for air pollution control. J. Chem. Technol. Biotechnol. 84, 1419–1436. https://doi.org/10.1002/jctb.2216.
Kumaraswamy, R., Kuenen, J.G., Kleerebezem, R., Van Loosdrecht, M.C.M., Muyzer, G., 2006. Structure of microbial communities performing the simultaneous reduction of Fe(II)EDTA.NO^{2-} and Fe(III)EDTA$^-$. Appl. Microbiol. Biotechnol. 73, 922–931. https://doi.org/10.1007/s00253-006-0542-z.
Kumaraswamy, R., Van Dongen, U., Kuenen, J.G., Abma, W., Van Loosdrecht, M.C.M., Muyzer, G., 2005. Characterization of microbial communities removing nitrogen oxides from flue gas: the BioDeNOx process. Appl. Environ. Microbiol. 71, 6345–6352. https://doi.org/10.1128/AEM.71.10.6345-6352.2005.
Lee, B.D., Apel, W.A., Smith, W.A., 2001. Oxygen effects on thermophilic microbial populations in biofilters treating nitric oxide containing off-gas streams. Environ. Prog. 20, 157–166. https://doi.org/10.1002/ep.670200311.
Lee, J.N., Lee, J.S., Shin, C.S., Park, S.C., Kim, S.W., 2000. Methods to enhance tolerances of *Chlorella* KR-1 to toxic compounds in flue gas. Appl. Biochem. Biotechnol. Part A Enzym. Eng. Biotechnol. 84– 86, 329–342. https://doi.org/10.1385/ABAB:84-86:1-9:329.
Lee, K.H., Sublette, K.L., 1991. Simultaneous combined microbial removal of sulfur dioxide and nitric oxide from a gas stream. Appl. Biochem. Biotechnol. 28 (29), 623–634. https://doi.org/10.1007/BF02922637.
Lee, K.H., Sublette, K.L., 1990. Reduction of nitric oxide to elemental nitrogen by *Thiobacillus Denitrificans* - scientific note. Appl. Biochem. Biotechnol. 24 (25), 441–445. https://doi.org/10.1007/BF02920268.
Li, W., Li, M., Zhang, L., Zhao, J., Xia, Y., Liu, N., Li, S., Zhang, S., 2016a. Enhanced NOx removal performance and microbial community shifts in an oxygen-resistance chemical absorption-biological reduction integrated system. Chem. Eng. J. 290, 185–192. https://doi.org/10.1016/j.cej.2016.01.044.
Li, W., Liu, N., Cai, L.L., Jiang, J.L., Chen, J.M., 2011. Reduction of Fe(III) chelated with citrate in an NOx scrubber solution by *Enterococcus* sp. FR-3. Bioresour. Technol. 102, 3049–3054. https://doi.org/10.1016/j.biortech.2010.10.036.
Li, W., Wu, C.Z., Shi, Y., 2006. Metal chelate absorption coupled with microbial reduction for the removal of NOx from flue gas. J. Chem. Technol. Biotechnol. 81, 306–311. https://doi.org/10.1002/jctb.1394.
Li, W., Wu, C.Z., Zhang, S.H., Shao, K., Shi, Y., 2007. Evaluation of microbial reduction of Fe(III)EDTA in a chemical absorption-biological reduction integrated NOx removal system. Environ. Sci. Technol. 41, 639–644. https://doi.org/10.1021/es061757e.
Li, W., Xia, Y., Zhao, J., Liu, N., Li, S., Zhang, S., 2015a. Generation, utilization, and transformation of cathode electrons for bioreduction of Fe(III)EDTA in a biofilm electrode reactor related to NOx removal from flue gas. Environ. Sci. Technol. 49, 4530–4535. https://doi.org/10.1021/es5058488.
Li, X-k, Xu, J-l, Guo, Y, Zhou, W-z, Yuan, Z-h, 2015b. Effects of simulated flue gas on components of *Scenedesmus raciborskii* WZKMT. Bioresour. Technol. 190, 339–344. https://doi.org/10.1016/j.biortech.2015.04.093.
Li, W., Zhang, L., Liu, N., Shi, Y., Xia, Y., Zhao, J., Li, M., 2014. Evaluation of NO removal from flue gas by a chemical absorption-biological reduction integrated system: complexed NO conversion pathways and nitrogen equilibrium analysis. Energy Fuel. 28, 4725–4730. https://doi.org/10.1021/ef500652g.
Li, W., Zhao, J., Zhang, L., Xia, Y., Liu, N., Li, S., Zhang, S., 2016b. Pathway of FeEDTA transformation and its impact on performance of NOx removal in a chemical absorption-biological reduction integrated process. Sci. Rep. 6, 2–11. https://doi.org/10.1038/srep18876.

Lin, T., Zhou, Z., Liu, Y., Wang, X., Jing, G., 2014. Evaluation of Fe(III)EDTA and Fe(II) EDTA-NO reduction in a NOx scrubber solution by magnetic Fe_3O_4-chitosan microspheres immobilized microorganisms. Biotechnol. Bioproc. Eng. 19, 175–182. https://doi.org/10.1007/s12257-013-0207-6.

Liu, B., Zhao, J., Wei, F., 2013. Characterization of caprolactam based eutectic ionic liquids and their application in SO_2 absorption. J. Mol. Liq. 180, 19–25. https://doi.org/10.1016/j.molliq.2012.12.024.

Liu, N., Jiang, Y., Zhang, L., Xia, Y., Lu, B., Xu, B., Li, W., Li, S., 2014. Evaluation of NOx removal from flue gas by a chemical absorption-biological reduction integrated system: glucose consumption and utilization pathways. Energy Fuel. 28, 7591–7598. https://doi.org/10.1021/ef5014852.

Liu, N., Li, Y. Ying, Ouyang, D. Juan, Guo, R., Chen, R., Li, W., Li, J., xiang, Zhao, hong, J., 2021. Study on NOx removal from simulated flue gas by an electrobiofilm reactor: EDTA-ferrous regeneration and biological kinetics mechanism. Environ. Sci. Pollut. Res. 28, 2860–2870. https://doi.org/10.1007/s11356-020-10617-2.

Liu, N., Lu, B.H., Zhang, S.H., Jiang, J.L., Cai, L.L., Li, W., He, Y., 2012. Evaluation of nitric oxide removal from simulated flue gas by Fe(II)EDTA/Fe(II)citrate mixed absorbents. Energy Fuel. 26, 4910–4916. https://doi.org/10.1021/ef300538x.

Lizzul, A.M., Hellier, P., Purton, S., Baganz, F., Ladommatos, N., Campos, L., 2014. Combined remediation and lipid production using *Chlorella sorokiniana* grown on wastewater and exhaust gases. Bioresour. Technol. 151, 12–18. https://doi.org/10.1016/j.biortech.2013.10.040.

Lockhart, H.B., Blakeley, R.V., 1975. Aerobic photodegradation of Fe(III)-(Ethylenedinitrilo)tetraacetate (ferric EDTA) implications for natural waters. Environ. Sci. Technol. 9, 1035–1038. https://doi.org/10.1021/es60110a009.

Long, X., Cao, li, xia, H., Duan, B., bei, Jia, lei, M., 2017. Removal of NO with the hexamminecobalt solution catalyzed by the carbon treated with oxalic acid. Environ. Sci. Pollut. Res. 24, 27788–27798. https://doi.org/10.1007/s11356-017-0328-0.

Long, X. li, Xin, Z. ling, Chen, M. bing, Li, W., Xiao, W. de, Yuan, W. Kang, 2008. Kinetics for the simultaneous removal of NO and SO_2 with cobalt ethylenediamine solution. Separ. Purif. Technol. 58, 328–334. https://doi.org/10.1016/j.seppur.2007.05.004.

Long, X., Xiao, W.-D., Yuan, W., 2005. Simultaneous absorption of NO and SO_2 into hexamminecobalt(II)/iodide solution. Chemosphere 59, 811–817. https://doi.org/10.1016/j.chemosphere.2004.11.005.

López-Archilla, A.I., Moreira, D., López-García, P., Guerrero, C., 2004. *Phytoplankton* diversity and *cyanobacterial* dominance in a hypereutrophic shallow lake with biologically produced alkaline pH. Extremophiles 8, 109–115. https://doi.org/10.1007/s00792-003-0369-9.

Lu, B.H., Jiang, Y., Cai, L.L., Liu, N., Zhang, S.H., Li, W., 2011. Enhanced biological removal of NOx from flue gas in a biofilter by Fe(II)Cit/Fe(II)EDTA absorption. Bioresour. Technol. 102, 7707–7712. https://doi.org/10.1016/j.biortech.2011.05.086.

Ma, S., Li, Da, Yu, Y., Li, Dianlin, Yadav, R.S., Feng, Y., 2019. Application of a microalga, *Scenedesmus obliquus* PF3, for the biological removal of nitric oxide (NO) and carbon dioxide. Environ. Pollut. 252, 344–351. https://doi.org/10.1016/j.envpol.2019.05.084.

Ma, S., Yu, Y., Cui, H., Li, J., Feng, Y., 2020. Utilization of domestic wastewater as a water source of *Tetradesmus obliquus* PF3 for the biological removal of nitric oxide. Environ. Pollut. 262, 114243 https://doi.org/10.1016/j.envpol.2020.114243.

Ma, S.H., Wen, Z.G., Chen, J.N., Wen, Z.C., 2014. Mode of circular economy in China's iron and steel industry: a case study in Wu'an city. J. Clean. Prod. 64, 505–512. https://doi.org/10.1016/j.jclepro.2013.10.008.

Manconi, I., van der Maas, P., Lens, P.N.L., 2006. Effect of sulfur compounds on biological reduction of nitric oxide in aqueous Fe(II)$EDTA^{2-}$ solutions. Nitric Oxide - Biol. Chem. 15, 40–49. https://doi.org/10.1016/j.niox.2005.11.012.

Matsumoto, H., Hamasaki, A., Sioji, N., Ikuta, Y., 1997. Influence of CO_2, SO_2 and no in flue gas on microalgae productivity. J. Chem. Eng. Jpn. https://doi.org/10.1252/jcej.30.620.

Mi, X.H., Gao, L., Zhang, S.H., Cai, L.L., Li, W., 2009. A new approach for Fe(III)EDTA reduction in NOx scrubber solution using bio-electro reactor. Bioresour. Technol. 100, 2940–2944. https://doi.org/10.1016/j.biortech.2009.01.046.

Min, K.N., Ergas, S.J., Harrison, J.M., 2002. Hollow-fiber membrane bioreactor for nitric oxide removal. Environ. Eng. Sci. 19, 575–583. https://doi.org/10.1089/109287502320963517.

Mladenović, M., Paprika, M., Marinković, A., 2018. Denitrification techniques for biomass combustion. Renew. Sustain. Energy Rev. 82, 3350–3364. https://doi.org/10.1016/j.rser.2017.10.054.

Nagase, H., Eguchi, K., Yoshihara, K.I., Hirata, K., Miyamoto, K., 1998. Improvement of microalgal NOx removal in bubble column and airlift reactors. J. Ferment. Bioeng. 86, 421–423. https://doi.org/10.1016/S0922-338X(99)89018-7.

Nagase, H., Yoshihara, K. ichi, Eguchi, K., Okamoto, Y., Murasaki, S., Yamashita, R., Hirata, K., Miyamoto, K., 2001. Uptake pathway and continuous removal of nitric oxide from flue gas using microalgae. Biochem. Eng. J. 7, 241–246. https://doi.org/10.1016/S1369-703X(00)00122-4.

Nagase, H., Yoshihara, K.I., Eguchi, K., Yokota, Y., Matsui, R., Hirata, K., Miyamoto, K., 1997. Characteristics of biological NOx removal from flue gas in a *Dunaliella tertiolecta* culture system. J. Ferment. Bioeng. 83, 461–465. https://doi.org/10.1016/S0922-338X(97)83001-2.

Negoro, M., Shioji, N., Miyamoto, K., Micira, Y., 1991. Growth of microalgae in high CO_2 gas and effects of SOX and NOX. Appl. Biochem. Biotechnol. 28 (29), 877–886. https://doi.org/10.1007/BF02922657.

Niu, H., Leung, D.Y.C., 2010. A review on the removal of nitrogen oxides from polluted flow by bioreactors. Environ. Rev. 18, 175–189. https://doi.org/10.1139/A10-007.

Niu, H., Leung, D.Y.C., Wong, C., Zhang, T., Chan, M., Leung, F.C.C., 2014. Nitric oxide removal by wastewater bacteria in a biotrickling filter. J. Environ. Sci. 26, 555–565. https://doi.org/10.1016/S1001-0742(13)60456-8.

Nymoen, H., van Velzen, D., Langenkamp, H., 1993. Absorption of NO in aqueous solutions of Fe(II)EDTA: determination of the equilibrium constant. Chem. Eng. Process. https://doi.org/10.1016/0255-2701(93)87002-C.

Okuno, K., Hirai, M., Sugiyama, M., Haruta, K., Shoda, M., 2000. Microbial removal of nitrogen monoxide (NO) under aerobic conditions. Biotechnol. Lett. 22, 77–79. https://doi.org/10.1023/A:1005624815889.

Pandey, R.A., Biswas, R., Chakrabarti, T., Devotta, S., 2005. Flue gas desulfurization: physicochemical and biotechnological approaches. Crit. Rev. Environ. Sci. Technol. 35, 571–622. https://doi.org/10.1080/10643380500326374.

Puckett, L.J., Teague, M.W., 1971. Production of H3O+·nH2O from NO+ precursor in no–H2O gas mixtures. J. Chem. Phys. 54, 2564–2572. https://doi.org/10.1063/1.1675213.

Qie, F., Zhu, J., Rong, J., Zong, B., 2019. Biological removal of nitrogen oxides by microalgae, a promising strategy from nitrogen oxides to protein production. Bioresour. Technol. 292, 122037 https://doi.org/10.1016/j.biortech.2019.122037.

Qin, L., Zhang, Y., Xu, Z., Zhang, G., 2018. Advanced membrane bioreactors systems: new materials and hybrid process design. Bioresour. Technol. 269, 476–488. https://doi.org/10.1016/j.biortech.2018.08.062.

Rahimi, S., Modin, O., Mijakovic, I., 2020. Technologies for biological removal and recovery of nitrogen from wastewater. Biotechnol. Adv. 43, 107570 https://doi.org/10.1016/j.biotechadv.2020.107570.

Razaviarani, V., Ruiz-Urigüen, M., Jaffé, P.R., 2019. Denitrification of nitric oxide using hollow fiber membrane bioreactor; effect of nitrate and nitric oxide loadings on the reactor performance and microbiology. Waste and Biomass Valorization 10, 1989. https://doi.org/10.1007/s12649-018-0223-z, 2000.

Rosenberger, S., Laabs, C., Lesjean, B., Gnirss, R., Amy, G., Jekel, M., Schrotter, J.-C., 2006. Impact of colloidal and soluble organic material on membrane performance in membrane bioreactors for municipal wastewater treatment. Water Res. 40, 710–720. https://doi.org/10.1016/j.watres.2005.11.028.

Sada, E., Kumazawa, H., Machida, H., 1987. Oxidation kinetics of FeII-EDTA and FeII-NTA chelates by dissolved oxygen. Ind. Eng. Chem. Res. 26, 1468–1472. https://doi.org/10.1021/ie00067a033.

Sakakibara, Y., Flora, J.R.V., Suidan, M.T., Kurodo, M., 1994. Modeling of electrochemically-activated denitrifying biofilms. Water Res. 28, 1077–1086. https://doi.org/10.1016/0043-1354(94)90193-7.

Sakakibara, Y., Nakayama, T., 2001. A novel multi-electrode system for electrolytic and biological water treatments. Water Res. 35, 768–778. https://doi.org/10.1016/s0043-1354(00)00327-4.

Sander, R., 2015. Compilation of Henry's law constants (version 4.0) for water as solvent. Atmos. Chem. Phys. 15, 4399–4981. https://doi.org/10.5194/acp-15-4399-2015.

Santiago, D.E.O., Jin, H.F., Lee, K., 2010. The influence of ferrous-complexed EDTA as a solubilization agent and its auto-regeneration on the removal of nitric oxide gas through the culture of green alga *Scenedesmus* sp. Process Biochem. 45, 1949–1953. https://doi.org/10.1016/j.procbio.2010.04.003.

Schmidt, I., Hermelink, C., Van de Pas-Schoonen, K., Strous, M., Op den Camp, H.J., Kuenen, J.G., Jetten, M.S.M., 2002. Anaerobic ammonia oxidation in the presence of nitrogen oxides (NOx) by two different lithotrophs. Appl. Environ. Microbiol. 68, 5351–5357. https://doi.org/10.1128/AEM.68.11.5351-5357.2002.

Schneppensieper, T., Finkler, S., Czap, A., van Eldik, R., Heus, M., Nieuwenhuizen, P., Wreesmann, C., Abma, W., 2001a. Tuning the reversible binding of NO to iron(II) aminocarboxylate and related complexes in aqueous solution. Eur. J. Inorg. Chem. 2001 491. https://doi.org/10.1002/1099-0682, 200102)2001:2<491::aid-ejic491>3.3.co;2-u.

Schneppensieper, T., Wanat, A., Stochel, G., Goldstein, S., Meyerstein, D., Van Eldik, R., 2002. Mechanistic information on the reversible binding of NO to selected iron (II) chelates from activation parameters. Inorg. Chem. 41, 2565–2573.

Schneppensieper, T., Wanat, A., Stochel, G., Goldstein, S., Meyerstein, D., Van Eldik, R., 2001b. Ligand effects on the kinetics of the reversible binding of NO to selected aminocarboxylato complexes of iron(II) in aqueous solution. Eur. J. Inorg. Chem. 2317–2325. https://doi.org/10.1002/1099-0682(200109)2001:9<2317::AID-EJIC2317>3.0.CO;2.

Shen, J., Cai, L., Mao, Z., Sha, H., 2022. Research progress and kinetic model review of biological processes for purifying SO_2 and NOx. Int. J. Environ. Sci. Technol. 19, 9267–9282. https://doi.org/10.1007/s13762-021-03668-6.

Skalska, K., Miller, J.S., Ledakowicz, S., 2010. Trends in NOx abatement: a review. Sci. Total Environ. 408, 3976–3989. https://doi.org/10.1016/j.scitotenv.2010.06.001.

Sørensen, J., Rasmussen, L.K., Koike, I., 1987. Micromolar sulfide concentrations alleviate acetylene blockage of nitrous oxide reduction by denitrifying Pseudomonas fluorescens. Can. J. Microbiol. 33, 1001–1005. https://doi.org/10.1139/m87-176.

Straub, K.L., Benz, M., Schink, B., 2001. Iron metabolism in anoxic environments at near neutral pH. FEMS Microbiol. Ecol. 34, 181–186. https://doi.org/10.1016/S0168-6496(00)00088-X.

Sun, Congcong, Yuan, J., Xu, H., Huang, S., Wen, X., Tong, N., Zhang, Y., 2019a. Simultaneous removal of nitric oxide and sulfur dioxide in a biofilter under micro-oxygen thermophilic conditions: removal performance, competitive relationship and bacterial community structure. Bioresour. Technol. 290, 121768 https://doi.org/10.1016/j.biortech.2019.121768.

Sun, C., Zhang, Y., 2018. Complexing absorption of NO by cobalt(II)-Histidine. Energy Fuel. 32, 688–695. https://doi.org/10.1021/acs.energyfuels.7b02951.

Sun, C., Zhang, Y., Qu, Z., Zhou, J., 2020a. Simultaneous cobalt(III)-histidine reduction and aerobic denitrification by *Paracoccus versutus* LYM. Bioresour. Technol. 310, 123404 https://doi.org/10.1016/j.biortech.2020.123404.

Sun, Chaoyue, Zhang, Y., Qu, Z., Zhou, J., 2019b. Effects of cobalt-histidine absorbent on aerobic denitrification by *Paracoccus versutus* LYM. Amb. Express 9. https://doi.org/10.1186/s13568-019-0927-x.

Sun, J., Zhang, Y., Dong, X., Chen, M., Zhou, J., 2015. Regeneration of spent NOx scrubber liquor using a dual-chamber microbial fuel cell. J. Chem. Technol. Biotechnol. 90, 1692–1698. https://doi.org/10.1002/jctb.4479.

Sun, Y., Gao, M., Ren, S., Zhang, Q., Hou, Y., Wu, W., 2020b. Highly efficient absorption of NO by amine-based functional deep eutectic solvents. Energy Fuel. 34, 690–697. https://doi.org/10.1021/acs.energyfuels.9b03335.

Sutton, H.C., 1985. Efficiency of chelated iron compounds as catalysts for the Haber-Weiss reaction. J. Free Radic. Biol. Med. 1, 195–202. https://doi.org/10.1016/0748-5514(85)90118-7.

Thomas, D., Vanderschnren, J., 2000. Nitrogen oxides scrubbing with alkaline solutions. Chem. Eng. Technol. 23, 449–455. https://doi.org/10.1002/(SICI)1521-4125, 200005)23:5<449::AID-CEAT449>3.0.CO;2-L.

Tucker, M.D., Barton, L.L., Thomson, B.M., Wagener, B.M., Aragon, A., 1999. Treatment of waste containing EDTA by chemical oxidation. Waste Manag. 19, 477–482. https://doi.org/10.1016/S0956-053X(99)00235-4.

Van Den Hende, S., Vervaeren, H., Boon, N., 2012. Flue gas compounds and microalgae: (Bio-)chemical interactions leading to biotechnological opportunities. Biotechnol. Adv. 30, 1405–1424. https://doi.org/10.1016/j.biotechadv.2012.02.015.

van der Maas, P., 2005. Chemically enhanced biological NOx removal from flue gases. In: Nitric oxide and ferric EDTA reduction in BioDeNOx reactors. Wageningen Universiteit.

van der Maas, P., Harmsen, L., Weelink, S., Klapwijk, B., Lens, P., 2004. Denitrification in aqueous FeEDTA solutions. J. Chem. Technol. Biotechnol. 79, 835–841. https://doi.org/10.1002/jctb.1057.

van der Maas, P., Manconi, I., Klapwijk, B., Lens, P., 2008. Nitric oxide reduction in BioDeNOx reactors: kinetics and mechanism. Biotechnol. Bioeng. 100, 1099–1107. https://doi.org/10.1002/bit.21841.

van der Maas, P., Peng, S., Klapwijk, B., Lens, P., 2005a. Enzymatic versus nonenzymatic conversions during the reduction of EDTA-chelated Fe(III) in BioDeNOx reactors. Environ. Sci. Technol. 39, 2616–2623. https://doi.org/10.1021/es049222d.

van der Maas, P., van de Sandt, T., Klapwijk, B., Lens, P., 2003. Biological reduction of nitric oxide in aqueous Fe(II)EDTA solutions. Biotechnol. Prog. 19, 1323–1328. https://doi.org/10.1021/bp030008n.

van der Maas, P., van den Bosch, P., Klapwijk, B., Lens, P., 2005b. NOx removal from flue gas by an integrated physicochemical absorption and biological denitrification process. Biotechnol. Bioeng. 90, 433–441. https://doi.org/10.1002/bit.20420.

van der Maas, P., van den Brink, P., Utomo, S., Klapwijk, B., Lens, P., 2006. NO removal in continuous BioDeNOx reactors: $Fe(II)EDTA^{2-}$ regeneration, biomass growth, and EDTA degradation. Biotechnol. Bioeng. 94, 575–584. https://doi.org/10.1002/bit.20859.

van der Mass, P., van den Brink, P., Klapwijk, B., Lens, P., 2009. Acceleration of the Fe (III)$EDTA^{-}$ reduction rate in BioDeNOx reactors by dosing electron mediating compounds. Chemosphere 75, 243–249. https://doi.org/10.1016/j.chemosphere.2008.04.043.

Van Eynde, E., Lenaerts, B., Tytgat, T., Blust, R., Lenaerts, S., 2016. Valorization of flue gas by combining photocatalytic gas pretreatment with microalgae production. Environ. Sci. Technol. 50, 2538–2545. https://doi.org/10.1021/acs.est.5b04824.

Wang, J., Wu, C., Chen, J., Zhang, H., 2006. Denitrification removal of nitric oxide in a rotating drum biofilter. Chem. Eng. J. 121, 45–49. https://doi.org/10.1016/j.cej.2006.04.004.

Wang, X., Xu, X., Liu, S., Zhang, Y., Zhao, C., Yang, F., 2016. Combination of complex adsorption and anammox for nitric oxide removal. J. Hazard Mater. 312, 175–183. https://doi.org/10.1016/j.jhazmat.2016.03.034.

Wang, X., Xu, X., Zou, Y., Yang, F., Zhang, Y., 2018. Nitric oxide removal from flue gas with ammonium using AnammoxDeNOx process and its application in municipal sewage treatment. Bioresour. Technol. 265, 170–179. https://doi.org/10.1016/j.biortech.2018.05.096.

Wang, Xiaojun, Zhang, Y., Dong, X., Chen, M., Shi, Z., Zhou, J., 2013. Fe(II)EDTA-NO reduction by sulfide in the anaerobic aqueous phase: stoichiometry and kinetics. Energy Fuel. 27, 6024–6030. https://doi.org/10.1021/ef401095f.

Wang, Xiaoyan, Zhou, Z., Jing, G., 2013. Synthesis of Fe_3O_4 poly(styrene-glycidyl methacrylate) magnetic porous microspheres and application in the immobilization of *Klebsiella* sp. FD-3 to reduce Fe(III)EDTA in a NOx scrubbing solution. Bioresour. Technol. 130, 750–756. https://doi.org/10.1016/j.biortech.2012.12.010.

Wang, X.C., Bi, X.Y., Sun, P.S., Chen, J.Q., Zou, P., Ma, X.M., Zhang, J., Wang, H.Y., Xu, X.Y., 2015. Effects of oxygen content on the simultaneous microbial removal of SO_2 and NOx in biotrickling towers. Biotechnol. Bioproc. Eng. 20, 924–930. https://doi.org/10.1007/s12257-015-0138-5.

Wang, Y., Ren, S., Hou, Y., Wu, W., 2021. Eutectic solvents. Eutectic Solvents 1–28. https://doi.org/10.3390/books978-3-0365-0023-2.

Wei, Z.S., Huang, Q.R., Wang, J.B., Huang, Z.S., Chen, Z.Y., Li, B.R., 2016a. Performance and mechanism of nitric oxide removal using a thermophilic membrane biofilm reactor. Fuel Process. Technol. 148, 217–223. https://doi.org/10.1016/j.fuproc.2016.03.003.

Wei, Z.S., Li, B.R., Wang, J.B., Huang, Z.S., He, Y.M., Chen, Z.Y., 2016b. Coupling membrane catalysis and biodegradation for nitric oxide removal in a novel hybird catalytic membrane biofilm reactor. Chem. Eng. J. 296, 154–161. https://doi.org/10.1016/j.cej.2016.03.125.

Wei, Z.S., Wang, J.B., Huang, Z.S., Xiao, X.L., Tang, M.R., Li, B.L., Zhang, X., 2019. Removal of nitric oxide from biomass combustion by thermophilic nitrification-aerobic denitrification combined with catalysis in membrane biofilm reactor. Biomass Bioenergy 126, 34–40. https://doi.org/10.1016/j.biombioe.2019.05.004.

Wen, Z., Shen, H., Li, Y., Wang, Z., Wang, G., Cen, K., 2019. Experimental study on the NOx removal by scrubbing with urea–H2O2 solution after NO partial preoxidation. Energy Fuel. 33, 6600–6605. https://doi.org/10.1021/acs.energyfuels.9b01184.

Winkelman, J.G.M., Gambardella, F., Heeres, H.J., 2007. A rate based reactor model for BiodeNOx absorber units. Chem. Eng. J. 133, 165–172. https://doi.org/10.1016/j.cej.2007.03.002.

Wolak, M., van Eldik, R., 2002. To be or not to be NO in coordination chemistry? A mechanistic approach. Coord. Chem. Rev. 230, 263–282. https://doi.org/10.1016/S0010-8545(01)00472-6.

World Health Organization (WHO), 2000. Nitrogen dioxide. In: Air Quality Guidelines for Europe, second ed., pp. 175–180.

World Health Organization (WHO), 1997. Nitrogen Oxides, Nitrogen Oxides, second ed. Environmental Health Criteria, p. 188.

Wu, L., Wang, L.K., Wei, W., Ni, B.J., 2022. Autotrophic denitrification of NO for effectively recovering N_2O through using thiosulfate as sole electron donor. Bioresour. Technol. 347 https://doi.org/10.1016/j.biortech.2022.126681.

Wubs, H.J., Beenackers, A.A.C.M., 1994. Kinetics of H_2S absorption into aqueous ferric solutions of edta and hedta. AIChE J. 40, 433–444. https://doi.org/10.1002/aic.690400307.

Xia, Y., Shi, Y., Zhou, Y., Liu, N., Li, W., Li, S., 2014. A new approach for NOx removal from flue gas using a biofilm electrode reactor coupled with chemical absorption. Energy Fuel. 28, 3332–3338. https://doi.org/10.1021/ef500604d.

Xia, Y.F., Lu, B.H., Liu, N., Chen, Q.L., Li, S.J., Li, W., 2013. NOx removal in chemical absorption-biological reduction integrated system: process rate and rate-limiting step. Bioresour. Technol. 149, 184–190. https://doi.org/10.1016/j.biortech.2013.09.056.

Xu, X., Chang, S.G., 2007. Removing nitric oxide from flue gas using iron(II) citrate chelate absorption with microbial regeneration. Chemosphere 67, 1628–1636. https://doi.org/10.1016/j.chemosphere.2006.11.015.

Xu, X.J., Wu, Y.N., Xiao, Q.Y., Xie, P., Ren, N.Q., Yuan, Y.X., Lee, D.J., Chen, C., 2022. Simultaneous removal of NOX and SO_2 from flue gas in an integrated FGD-CABR system by sulfur cycling-mediated Fe(II)EDTA regeneration. Environ. Res. 205, 112541 https://doi.org/10.1016/j.envres.2021.112541.

Yang, W.F., Hsing, H.J., Yang, Y.C., Shyng, J.Y., 2007. The effects of selected parameters on the nitric oxide removal by biofilter. J. Hazard Mater. 148, 653–659. https://doi.org/10.1016/j.jhazmat.2007.03.023.

Yang, Y., Huang, S., Liang, W., Zhang, Y., Huang, H., Xu, F., 2012. Microbial removal of NO X at high temperature by a novel aerobic strain *Chelatococcus daeguensis* TAD1 in a biotrickling filter. J. Hazard Mater. 203 (204), 326–332. https://doi.org/10.1016/j.jhazmat.2011.12.031.

Yen, H.W., Ho, S.H., Chen, C.Y., Chang, J.S., 2015. CO_2, NOx and SOx removal from flue gas via microalgae cultivation: a critical review. Biotechnol. J. 10, 829–839. https://doi.org/10.1002/biot.201400707.

Yoshihara, K.I., Nagase, H., Eguchi, K., Hirata, K., Miyamoto, K., 1996. Biological elimination of nitric oxide and carbon dioxide from flue gas by marine microalga NOA-113 cultivated in a long tubular photobioreactor. J. Ferment. Bioeng. 82, 351–354. https://doi.org/10.1016/0922-338X(96)89149-5.

Zhang, C., Zhao, J., Sun, C., Li, S., Zhang, D., Guo, T., Li, W., 2018. Two-stage chemical absorption-biological reduction system for NO removal: system start-up and optimal operation mode. Energy Fuel. 32, 7701–7707. https://doi.org/10.1021/acs.energyfuels.8b00756.

Zhang, D., Ren, L., Yao, Z., Wan, X., Lu, P., Zhang, X., 2017. Removal of nitrogen oxide based on anammox through Fe(II)EDTA absorption. Energy Fuel. 31, 7247–7255. https://doi.org/10.1021/acs.energyfuels.7b00119.

Zhang, J., Ren, L., Zhang, D., Li, J., Peng, S., Han, X., Ding, A., Lu, P., 2019a. Reduction of NO to N_2 in an autotrophic up-flow bioreactor with sponge iron bed based Fe(II) EDTA complexation. Fuel 254, 115631. https://doi.org/10.1016/j.fuel.2019.115631.

Zhang, S.H., Cai, L.L., Mi, X.H., Jiang, J.L., Li, W., 2008a. NOx removal from simulated flue gas by chemical absorption-biological reduction integrated approach in a biofilter. Environ. Sci. Technol. 42, 3814–3820. https://doi.org/10.1021/es800200g.

Zhang, S.H., Li, W., Wu, C.Z., Chen, H., Shi, Y., 2007. Reduction of Fe(II)EDTA-NO by a newly isolated *Pseudomonas* sp. strain DN-2 in NOx scrubber solution. Appl. Microbiol. Biotechnol. 76, 1181–1187. https://doi.org/10.1007/s00253-007-1078-6.

Zhang, S.H., Mi, X.H., Cai, L.L., Jiang, J.L., Li, W., 2008b. Evaluation of complexed NO reduction mechanism in a chemical absorption-biological reduction integrated NOx removal system. Appl. Microbiol. Biotechnol. 79, 537–544. https://doi.org/10.1007/s00253-008-1469-3.

Zhang, S.H., Shi, Y., Li, W., 2012. Biological and chemical interaction of oxygen on the reduction of Fe(III)EDTA in a chemical absorption-biological reduction integrated NOx removal system. Appl. Microbiol. Biotechnol. 93, 2653–2659. https://doi.org/10.1007/s00253-011-3573-z.

Zhang, X., Jin, R., Liu, G., Dong, X., Zhou, J., Wang, A., 2013. Removal of nitric oxide from simulated flue gas via denitrification in a hollow-fiber membrane bioreactor. J. Environ. Sci. (China) 25, 2239–2246. https://doi.org/10.1016/S1001-0742(12)60285-X.

Zhang, X., Yue, F., Li, H., Huang, Y., Zhang, Y., Wen, H., Wang, J., 2016. Reversible oxygenation of α -amino acid–cobalt(II) complexes. Bioinorgan. Chem. Appl. 2016, 1–10. https://doi.org/10.1155/2016/3585781.

Zhang, Y., Sun, L., Zhou, J., 2019b. Simultaneous biological and chemical removal of sulfate and FE(II)EDTA-NO-no in anaerobic conditions and regulation of sulfate reduction products. Minerals 9. https://doi.org/10.3390/min9060330.

Zhao, J., Zhang, C., Li, M., Li, S., Li, W., Zhang, S., 2017. Two-stage chemical absorption-biological reduction system for NO removal: model development and footprint estimation. Energy Fuel. 31, 8454–8461. https://doi.org/10.1021/acs.energyfuels.7b01620.

Zhao, J., Zhang, C., Sun, C., Li, W., Zhang, S., Li, S., Zhang, D., 2018. Electron transfer mechanism of biocathode in a bioelectrochemical system coupled with chemical absorption for NO removal. Bioresour. Technol. 254, 16–22. https://doi.org/10.1016/j.biortech.2018.01.066.

Zheng, M., Li, C., Liu, S., Gui, M., Ni, J., 2016. Potential application of aerobic denitrifying bacterium *Pseudomonas aeruginosa* PCN-2 in nitrogen oxides (NOx) removal from flue gas. J. Hazard Mater. 318, 571–578. https://doi.org/10.1016/j.jhazmat.2016.07.047.

Zhou, Y., Gao, L., Xia, Y.F., Li, W., 2012. Enhanced reduction of Fe(II)EDTA-NO/Fe(III) EDTA in NOx scrubber solution using a three-dimensional biofilm-electrode reactor. Environ. Sci. Technol. 46, 12640–12647. https://doi.org/10.1021/es3025726.

Zhou, Z., Jing, G., Zheng, X., 2013a. Reduction of Fe(III)EDTA by *Klebsiella* sp. strain FD-3 in NOx scrubber solutions. Bioresour. Technol. 132, 210–216. https://doi.org/10.1016/j.biortech.2013.01.022.

Zhou, Z., Jing, G., Zhou, Q., 2013b. Enhanced NOx removal from flue gas by an integrated process of chemical absorption coupled with two-stage biological reduction using immobilized microorganisms. Process Saf. Environ. Protect. 91, 325–332. https://doi.org/10.1016/j.psep.2012.08.001.

Zielke, C.W., Wu, M.-C.M., 1986. Method of Reducing Sulfur Oxide and Nitrogen Oxide Conten of Flue Gas.

Zumft, W.G., 1997. Cell biology and molecular basis of denitrification. Microbiol. Mol. Biol. Rev. 61, 533–616. https://doi.org/10.1128/.61.4.533-616.1997.