

Comparison between Cyrene and NMP as Solvents for CO₂ Removal

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N-Methyl-2-Pyrrolidone (NMP) is one of the most used solvents for CO₂ removal from gaseous streams by physical absorption and, recently, it has also been considered as a component of hybrid solvents performing both physical and chemical absorption for possible applications to the capture of CO₂ from flue gas streams. However, NMP has some drawbacks and it has been included in the list of 'Substances of Very High Concern' according to the European Regulation No. 1907/2006. For this reason, alternative solvents are being considered. Cyrene is a biodegradable, non-mutagenic, non-toxic and biomass-derived solvent, having similar characteristics to NMP. The main similarity between the two species consists in the fact that they result to be dipolar aprotic solvents, as proved by the values of both Kamlet–Abboud–Taft and Hansen space parameters. Despite that, NMP and Cyrene differ in physical properties as boiling point, flash point, autoignition temperature and water solubility. However, to the authors' knowledge, no comparison is available in the literature regarding the solubility of CO₂, which is a key parameter for defining the suitability of a solvent for CO₂ removal.

In the context of the PRIN 2022 project "GREEN-based water-lean SOLvent for CO₂ capture" (GREENSOL), funded by the European Union – NextGenerationEU, a detailed analysis of all the CO₂ solubility data available in the literature is presented in this work in order to determine whether Cyrene can be used as a bio-based alternative to NMP as solvent for physical absorption.

1. Introduction

N-Methyl-2-pyrrolidone (NMP) is a highly polar molecule that exists as a liquid at room temperature (Basma et al., 2018). Based on the raw materials used, several processes for NMP production have been developed. However, the only method currently used in industry is the condensation of γ -butyrolactone with methylamine (Chemical Book, 2024). NMP primary application is as a solvent for CO₂ removal due to its low vapor pressure, low viscosity, high CO₂ absorption capacity, complete solubility in water and low corrosiveness (Usula et al., 2014; Eskandari et al., 2022). Key industrial uses of this physical solvent include hydrogen production from syngas and natural gas treatment, as reported by Murrieta-Guevara and Trejo Rodriguez (1984) and by Shokouhi et al. (2021). Specifically, NMP is employed in the Purisol[®] process licensed by Lurgi (Hochgesand, 1970), which can operate at ambient temperature or, when refrigerated, down to 258.15 K (Kriebel, 1989). However, NMP presents certain drawbacks, which have led to its inclusion in the list of 'Substances of Very High Concern' (SVHC) according to the European Regulation No. 1907/2006 "Registration, Evaluation, Authorisation and restriction of Chemicals" (REACH) (Jin et al., 2017). Consequently, alternative solvents are under consideration.

Cyrene (dihydrolevoglucosenone) is a biodegradable, non-mutagenic and non-toxic solvent derived from biomass, which has emerged as a promising bio-based alternative to NMP. It is a dipolar aprotic solvent produced through a two-step process, in particular cellulose-containing substrates are first pyrolyzed to produce levoglucosenone, followed by catalytic hydrogenation to obtain Cyrene (Kong and Dolzhenko, 2022). Due to its similar properties to NMP, several studies in the literature suggest Cyrene as a potential bio-based substitute. For example, Zhou et al. (2021) have proposed this substitution for its use in cathode processing for lithium

batteries and Fernandes et al. (2022) have analysed this replacement for liquid-phase exfoliation of graphite. Milescu (2021) investigated Cyrene as a replacement for polar aprotic solvents in applications such as polymer dissolution for graffiti removal, flavonoid extraction, carbon nanotube dispersion, poly(amide-imide) wire enamel production, and filtration membrane fabrication. This comparison had also been carried out previously by Sherwood et al. (2014) in the context of fluorination reaction and Menshutkin reaction and by Alves Costa Pacheco et al. (2016) in relation to fluorination reaction and Heck reaction.

Although Cyrene has been investigated as a potential alternative to NMP across several fields, to the authors' knowledge, no studies have examined its use specifically for CO₂ removal and no comparative studies with NMP have been found. Therefore, this work aims at presenting a detailed comparison of Cyrene and NMP by analyzing their physical properties, toxicological indices and solubility data to evaluate the Cyrene viability as a bio-based solvent alternative to NMP for the CO₂ physical absorption process.

2. Comparison

The main similarity between Cyrene and NMP concern their characteristics in being dipolar aprotic solvents, as proved by the values of both Kamlet–Abboud–Taft (KAT) and Hansen solubility parameters (HSPs).

The KAT parameters, also known as solvatochromic parameters, have been developed by M.J. Kamlet, J.L.M. Abboud and R.W. Taft (Kamlet and Taft, 1976; Kamlet et al., 1981) to describe the attractive solute-solvent interactions and they are quantitatively measured by three parameters: α , β and π^* (Taft et al., 1985). Specifically, α is a measure of the hydrogen-bonding capabilities of a solvent's hydrogen bond donor acidity; β is a measure of the hydrogen-bonding capabilities of a solvent's hydrogen bond acceptor basicity; π^* is a measure of the solvent's dipolarity/polarizability (Taft et al., 1985; Ranjesh et al., 2018).

The Hansen solubility parameters (HSPs), developed by Charles Hansen in 1967, predict molecular affinities, solubility and solubility-related phenomena by assuming that the solubility depends on three types of molecular interactions: dispersion forces (δ_d), polar forces (δ_p) and hydrogen bonding (δ_h) (Hansen, 1967). The dispersion forces (δ_d), associated to van der Waals interactions, arise from temporary electronic fluctuations and are typical of non-polar interactions (Hansen, 2007). The polar forces (δ_p) represent the dipole-dipole attraction forces, that are more intense than the dispersion forces, and they are characteristic of molecules with permanent dipoles (Hansen, 2007). The hydrogen bonding interactions (δ_h) correspond to specific interactions linked to hydrogen bonds (Hansen, 2007). In the Hansen model, the HSPs of a solvent can be represented as a vector with components δ_d , δ_p and δ_h and each of this component is a coordinate that allows the solvent to be located in a fixed point of a three-dimensional space called Hansen space (Hansen, 1967). The closer two solvents are in the Hansen space, the more likely they are to exhibit the same solubilizing properties (Sherwood et al., 2014). The similarity of Cyrene parameter π^* to that of highly polar solvents (Table 1) supports its classification as a viable alternative to traditional polar solvents, such as NMP (Sherwood et al., 2014; Alves Costa Pacheco et al., 2016). Additionally, the comparable values of their Hansen solubility parameters (Table 1) place Cyrene and NMP near each other in Hansen space, indicating they will have similar distances from a given solute and, consequently, comparable solubility profiles for that solute (Alves Costa Pacheco et al., 2016). This confirms the similarity of the two components according to the literature and the consideration of Cyrene as a potential substitute for NMP (Sherwood et al., 2014; Alves Costa Pacheco et al., 2016). To better understand all the characteristics of the two molecules, the following sections will present a comparison of their physical properties, toxicological indices and CO₂ solubility.

Table 1: Comparison between the Kamlet–Abboud–Taft and Hansen solubility parameters of Cyrene and NMP (Sherwood et al., 2014)

Solvent	α	β	π^*	δ_d [MPa ^{0.5}]	δ_p [MPa ^{0.5}]	δ_h [MPa ^{0.5}]
Cyrene	0.00	0.61	0.93	18.8	10.6	6.9
NMP	0.00	0.75	0.90	18.0	12.3	7.2

2.1 Physical properties

Table 2 lists all the physical properties considered in this study to assess whether Cyrene could serve as a substitute for NMP in CO₂ removal through physical absorption. Regarding their chemical structure, Cyrene is an optically active ketone composed of two fused rings, which form a cyclic acetal (Stini et al., 2022), while NMP is a nonaromatic molecule with a five-membered ring structure, a heteroatom and a methyl group (Basma et al., 2018). As for the melting point, Cyrene presents a higher value than NMP. However, both the solvents remain in the liquid state under ambient conditions as well as within the typical operating temperature range of an absorption column (303.15 – 323.15 K) and of the regeneration section, that is usually operated at higher temperature. Cyrene and NMP have comparable normal boiling points, with no significant difference between

their two values. In contrast, Cyrene exhibits a significantly lower vapor pressure compared to NMP. This property is a significant factor, as a suitable physical (or chemical) solvent must be non-volatile to minimize solvent losses due to evaporation during the absorption process (Kohl and Nielsen, 1997), which may need further treating of the gas and an additional make-up. Therefore, Cyrene's lower vapor pressure makes it superior to NMP in meeting this criterion, enhancing its suitability as a bio-based alternative solvent. Moreover, both the flash point and the autoignition temperature of Cyrene are higher than those of NMP, which is advantageous as it allows the use of higher temperatures in the solvent regeneration section without the risk of solvent degradation or the formation of flammable mixtures. In the context of viscosity, a lower value is generally preferred to facilitate efficient mass transfer rates (Kohl and Nielsen, 1997). Although Cyrene exhibits a higher viscosity compared to NMP, this does not compromise its suitability as a physical solvent for CO₂ removal, as its viscosity remains within an acceptable range. In fact, its viscosity is lower than the one of Poly(ethylene glycol) dimethyl ether, another traditional solvent widely used for the same purpose. The density values are quite similar for both solvents. The Cyrene hydro solubility is significantly higher than the one of NMP. This high-water solubility could be a problem, in particular for the gases characterized by a substantial water content, which, if not removed at least in bulk amount in a pretreatment section, may reduce the CO₂ absorption efficiency.

Table 2: Comparison between the physical properties of Cyrene and NMP (SDS stands for Safety DataSheet)

Physical property	Unit	Cyrene	NMP	Measured at	Source
Molecular weight	[g/mol]	128.13	99.13		(SDS - Cyrene; SDS - NMP)
Melting point	[K]	< 253.15	249.15		(SDS - Cyrene; SDS - NMP)
Normal boiling point	[K]	479.70	477.23	101325 Pa	Aspen Plus® V14
Vapor pressure	[Pa]	7.49	45.95	298.15 K	Aspen Plus® V14
Flash point	[K]	381.15	364.15		(SDS - Cyrene; SDS - NMP)
Autoignition temperature	[K]	569.15	518.15	1013, 1.013 hPa	(SDS - Cyrene; SDS - NMP)
Dynamic viscosity	[cP]	4.80	1.89	298.15 K, 101325 Pa	Aspen Plus® V14
Density	[g/mL]	1.25	1.03	298.15 K	Aspen Plus® V14
Hydro solubility	[g/l]	52.6	1.00	293.15 K	(SDS - Cyrene, SDS - NMP)

2.2 Toxicological index

Considering that in 2011 NMP has been included in the SVHC list due to its classification as toxic for reproduction (ECHA, 2011), and given that Cyrene has been proposed as a safer bio-based alternative, as reported by Sherwood et al. (2014) and by Alves Costa Pacheco et al. (2016), this study also focuses on the analysis of additional toxicity indicators for both the substances. The toxicological indices listed in Table 3 have been estimated by using the Toxicity Estimation Software Tool (T.E.S.T.), that is based on Quantitative Structure Activity Relationships (QSARs) (Martin, 2021). These QSARs are mathematical models that predict toxicity based on the physical characteristics of the chemicals structure (Martin, 2021). Among the QSAR approaches available in T.E.S.T., the consensus method has been selected in this study for its superior prediction accuracy, as it generates estimates by averaging the toxicity predictions from all available QSAR models (Martin, 2021). The toxicological indices analyzed in this work are: 96-hour fathead minnow (LC50), oral rat (LD50), bioconcentration factor (BCF) and Ames mutagenicity. In particular:

- the LC50 value refers to the concentration of a chemical in water that results in the mortality of 50% of fathead minnows over a four-day exposure period;
- the LD50 endpoint denotes the dose of a chemical, expressed as mass of the chemical per body weight of the rat, that causes the death of 50% of rats following oral ingestion;
- the bioconcentration factor (BCF) is defined as the ratio between the chemical concentration in an organism and the chemical concentration in the surrounding water, achieved through absorption across respiratory surfaces, under steady-state conditions;
- the Ames test detects frame-shift mutations or base-pair substitutions by exposing histidine-dependent strains of *Salmonella typhimurium* to the chemical under evaluation. Mutagenic compounds induce reverse mutations that restore the bacteria's ability to synthesize histidine, enabling colony growth on a medium deficient in histidine.

By comparing the toxicity indices with their respective limit values reported in Table 3, it is possible to conclude that Cyrene and NMP fall within the same toxicity categories for all parameters except LD50, where only Cyrene can be classified as non-toxic.

Table 3: Toxicological indices for of Cyrene and NMP

Toxicological indices	Unit	Cyrene	NMP	Limit values
LC50	[mg/l]	3340.77	774.51	> 100: practically nontoxic (EPA, 2024)
LD50	[mg/kg]	5906.25	1846.39	500 – 2000: slightly toxic > 2000: nontoxic (EPA, 2024)
BCF	[-]	2.49	1.02	< 1000: no bioaccumulation (EHAC, 2012)
Ames mutagenicity	[-]	Negative	Negative	

2.3 CO₂ solubility

The primary factor influencing the CO₂ physical absorption in a solvent is the solubility of CO₂ in the liquid, which generally increases as the CO₂ partial pressure increases and decreases as the temperature decreases (Ban et al., 2014). Therefore, the optimal conditions for the CO₂ absorption process are typically high CO₂ partial pressures and low temperatures (Kohl and Nielsen, 1997). In the available literature, only two sources report the CO₂ solubility data for Cyrene. Kerelaux et al. (2022) provides eight points of solubility data at 313.15 K for pressures up to 4 MPa and Hajlaoui et al. (2023) reports only a single data point at 333.15 K and 4 MPa. Considering that the absorption columns generally operate within a temperature range from 303.15 to 313.15 K, this study has focused on the comparison of the CO₂ solubility in NMP and in Cyrene within this temperature range and the experimental points of Kerleaux et al. (2022) have been taken into account. The considered experimental data of CO₂ solubility in NMP are reported in Rajasingam et al. (2004), Bohloul et al. (2014) and Shokouhi et al. (2021). With these data, a comparative analysis between Cyrene and NMP is not feasible, as data at temperatures between 303.15 K and 313.15 K are missing, and those at 313.15 K are at a different pressure than the available experimental data for NMP. Therefore, to enable a meaningful comparison, the CO₂ solubility in Cyrene has been simulated by using the COnductor-like Screening MOdel for Real Solvents (COSMO-RS). This model, developed by Klamt in 1995, employs a statistical-thermodynamic approach grounded in quantum chemistry to predict the thermodynamic properties of fluids without requiring experimental input (Klamt, 1995). To verify the model accuracy, the CO₂ solubility in Cyrene at 313.15 K and pressures up to 4 MPa has been calculated via COSMO-RS and, then, compared to the experimental data provided by Kerelaux et al. (2022) (Figure 2). Higher deviations are observed at CO₂ partial pressures higher than 2.5 MPa (conditions not of interest for the purposes of this work). However, with an Average Absolute Deviation percentage (AAD%) of 14.07%, the prediction of the model can be considered acceptable for the target temperature and pressure ranges where experimental data are lacking. The CO₂ solubility in Cyrene has been calculated at 303.15 K over the CO₂ partial pressure range from 0.121 to 0.801 MPa, at 308.2 K from 0.96 to 6.67 MPa and at 313.15 K from 0.135 to 1.12 MPa. These pressure ranges have been selected to align with the conditions for which experimental data on NMP are available, enabling a direct comparison. Figure 2 presents the calculated solubility values for Cyrene in comparison with the experimental solubility data for NMP. The results demonstrate a similar solubility profile between Cyrene and NMP, suggesting that Cyrene could be a viable alternative solvent to NMP for CO₂ removal through physical absorption processes.

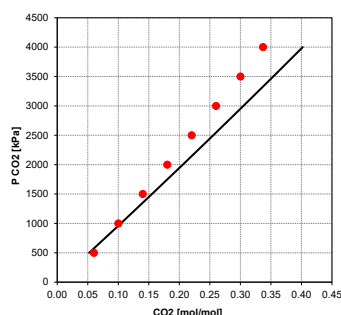


Figure 1. Comparison between CO₂ solubility experimental data provided by Kerleaux et al. (2022) (●) and CO₂ solubility profile calculated via COSMO-RS (—).

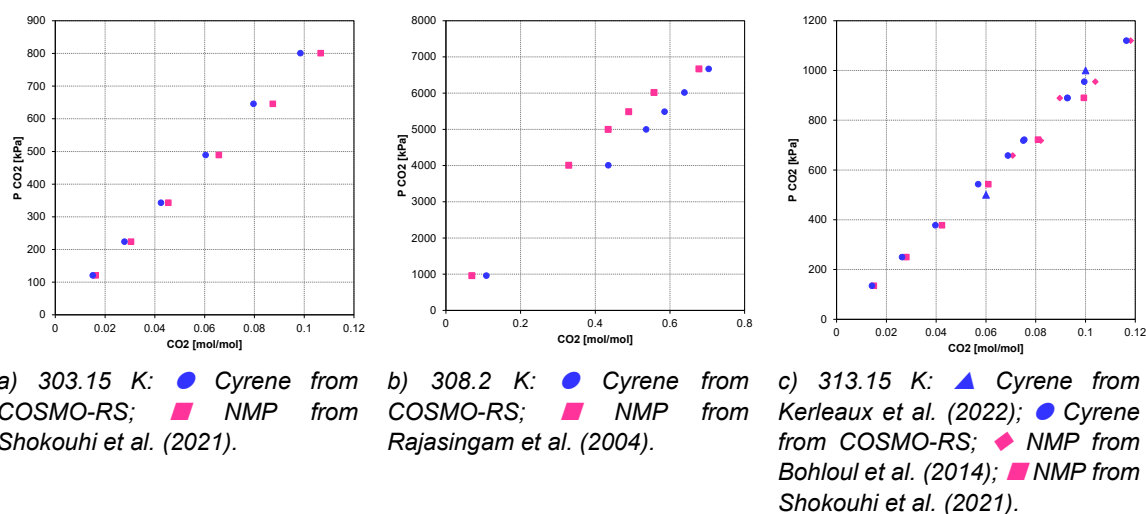


Figure 2. Comparison between CO_2 solubility in NMP and Cyrene at: a) 303.15 K; b) 308.2 K; c) 313.15 K.

3. Conclusions

This study is the first one in the literature, to the authors' knowledge, presenting a detailed comparative analysis between Cyrene and NMP, focusing on the potential of Cyrene as a bio-based alternative solvent to the toxic NMP physical absorption process. Given that several studies mention Cyrene as a replacement for NMP, this work examines its viability in applications related to CO_2 removal, such as hydrogen production from syngas streams and natural gas treatment. The analysis considers physical properties, toxicological indices and solubility data. As for the physical properties, the main distinction between the two molecules lies in their molecular structure. As for the toxicological profiles, NMP is toxic in terms of both reproductive impact and Lethal Dose 50, whereas Cyrene does not exhibit these toxic properties. Finally, in terms of solubility, a comparison of the experimental CO_2 solubility data for NMP with calculated data for Cyrene shows that the two molecules are characterized by a similar solubility profile at the typical conditions of the absorption section. In conclusion, Cyrene could be considered as a possible bio-based alternative solvent for CO_2 removal and is expected to align well with the NMP performance in CO_2 physical absorption applications.

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