

Numerical numerical model for the separation of carbon dioxide from natural gases

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ABSTRACT:

The increase in environmental temperatures and the transition of nature to two seasons, requires the making of some crucial decisions to ensure sustainability in the future.

That is precisely why the European Union, through the Net-Zero Directive, proposed accelerating the transition towards climate neutrality, by imposing the elimination of the use of fossil fuels.

For the transition to the use of energy fuels with CO₂ emissions reduced to zero, the use of natural gas was accepted as a transitional fuel.

Under these conditions, the consumption of natural gas will increase in the future, as it is necessary to exploit condensate and impure gas deposits.

The article proposes techniques for separating carbon dioxide by supersonic methods.

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I. INTRODUCTION

Currently, the methods for removing CO₂ from natural gases include chemical adsorption processes, physical adsorption, condensation and the use of separation membranes [1,2,3].

The need to remove carbon dioxide from natural gas is due to:

The possibility of the formation of gas hydrates (carbonic ice) in the transport pipelines,

Reducing their caloric capacity,

The risk in the exploitation and use of natural gas in chemical and electricity production facilities.

Classical methods of carbon dioxide removal are energy-consuming and also require chemical products that must be subsequently reprocessed.

Due to the need to improve energy efficiency and environmental conservation (protection), the use of supersonic separators has been proposed.

Through the analysis carried out in this chapter, We will describe the use of supersonic technology in the removal of carbon dioxide from extracted natural gases and also present a complex numerical model based on the use of artificial intelligence regarding the separation stages of of this gas (carbon dioxide), in order to capture and store it.

Clean natural gas leaves through the top of the absorber, while CO₂ solvent leaves the bottom of the absorber.

The vapors from the first unit are recompressed and mixed with the natural gas flow. The vapor from the second unit, which comprises mainly CO₂, forms the CO₂ product stream, which can then be stored or used for enhanced recovery.

The clean solvent is re-compressed and re-injected into the absorbent.

II. ONEW METHOD OF AMINE SOLVENT RECOVERY WITH ACID ADDITION

Chemical absorption technologies using a solvent have many advantages for carbon dioxide capture in the amine-based post-combustion capture system.

An alternative stripping process has been studied using an organic acid to decrease the solubility of carbon dioxide [4].

This method of amine recovery is new and is carried out in two stages of the crystallization process to separate the amine and the acid.

The proposed method involves the addition of an organic acid solution, in order to easily separate CO₂ from the amine, and to reduce the regeneration energy of the absorbent.

In this process, part of the absorption process is similar to that of the MEA absorption process but the regeneration part of the absorbent is improved to reduce energy costs.

The process is shown in Fig. 1.

The process to regenerate the chemical solvent has three parts:

- removing CO₂ from the amine solvent by adding an acid,
- separating the solvent by reacting the organic acid with a strong base,
- acid recovery by the reaction between an organic acid salt and a strong acid.

First, the CO₂-rich solvent leaves the absorber and enters the stripper.

The contact of the aqueous acid solution in the stripper and the organic acid solution changes the pH of the CO₂-rich solvent.

Changing the pH decreases the CO₂ solubility of the solvent and eliminates CO₂ in the gas phase.

The organic acid reacts with the carbamate, which has a high reaction enthalpy.

Thus, the addition of acid can reduce the recovery energy of the solvent by increasing the solubility of CO₂.

Thus, the addition of acid can reduce the operating temperature of the stripper and could thus be decreased from 383.15-403.15 K to approximately 313.15-353.15 K.

The stripper stream consists of an amine solution and an acid solution.

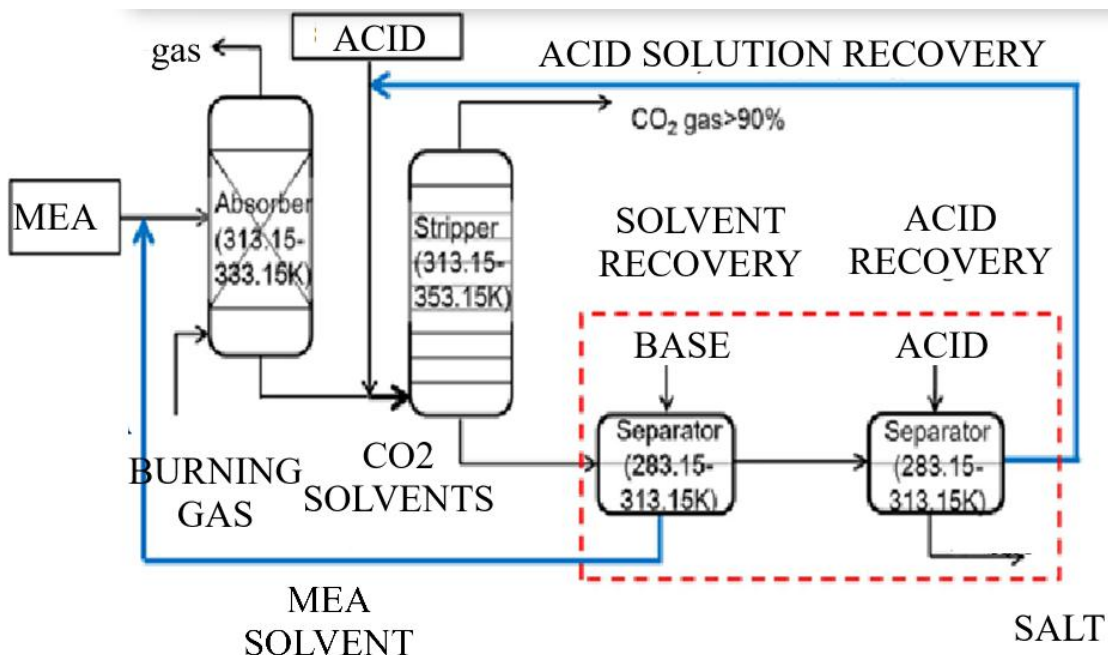


Figure 1. Flow diagram of the CO₂ absorption process[4]

Second, the aqueous amine and acid solution are separated by adding a strong base in a cooling crystallizer.

The organic acid reacts with a strong base (eg sodium hydroxide) and is converted into the salt of the organic acid.

The salt then precipitates due to the low solubility of the salts in the mixture. While the recovery of amines is based on the high pressure distillation process, this method uses the crystallization reaction which requires much less energy.

Finally, the crystallized product, the organic acid salt (sodium oxalate) is reacted with a strong acid such as hydrochloric acid, and the acid is recovered as crystals by precipitation at about 283.15-313.15 K.

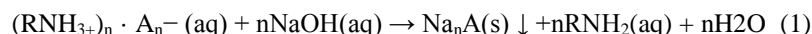
Then, the recovered organic acid is recycled to the stripper and the aqueous salt solution (NaCl) will be treated.

The reaction between two complex organic compounds or between an acid and a base forms a dissolved intermediate as a molecule or salt, then the intermediate is converted into a desired solid. This process changes the chemical balances in a mixture and makes the desired compound easily isolated.

In the MEA solvent recovery process after the acid addition separation process, the main reaction is based on equation 1.

The acid salt forms a solid phase, due to the low solubility of the salts in the amine-based solution, and is separated from the solvent.

When the amine solvent is activated, its absorption capacity increases.



In the acid recovery process, after the MEA solvent recovery process, the main reaction is defined by 2. The acid salt can convert to the acid by adding a strong acid such as hydrochloric acid.

III. CO₂ absorption in aqueous solutions of ionic liquids and MDEA

In recent years, ionic liquids have received increased attention from the scientific and industrial community [4].

They are liquid at temperatures close to or even well below room temperature, and are made up entirely of ions, resulting in extremely low volatility.

Their properties can be tailored to a certain extent by judicious selection of constituent ions in order to meet the necessary requirements for a particular application.

In addition, they usually exhibit fairly good thermal and chemical stability, as well as a high ability to dissolve a wide variety of compounds. In terms of gas solubility, carbon dioxide has been found to be much more soluble in many ionic liquids than other gases such as nitrogen or oxygen.

Together with the described set of physicochemical properties, this preferential solubility has led to the consideration of ionic liquids as a potential basis of an alternative technology for CO₂ capture in industrial applications.

Currently, the most used technology in CO₂ capture is absorption with aqueous amine solutions, which can capture CO₂ in stoichiometric ratios of 1:2 or 1:1, depending on the type of amine used.

However, a series of problems such as chemical degradation, emissions produced by evaporation, and high energy consumption (especially related to desorption, which requires temperatures in the range 373 - 413 K), are the main disadvantages of these processes, which urges researchers to look for better alternatives. Ionic liquids can be the basis for one of these alternatives.

Numerous ionic liquids have been tested to date for their CO₂ absorption capacity. Chemisorption is a mechanism to lead to a higher gas absorption capacity (more comparable to currently used CO₂ capture technologies), but the covalent bonds established between it and the ionic liquid complicate the desorption process for CO₂ recovery.

Ionic liquids are liquids that contain only ions.

The term ionic liquid is limited to substances that are liquid salts with a melting temperature below 1000 C.

Typically they are organic salts or eutectic mixtures of organic salt and inorganic salt.

Ionic liquids that are liquid at room temperature (T=250C) are called RTIL ("room-temperature ionic liquids").

Operating conditions: Absorber: T ~ 40 ° C, P ~ 1 atm, Stripper: T ~ 140 ° C, P > 1 atm

Ionic liquids (ILs) are a recent introduction to CO₂ capture as a potential replacement for traditional amines.

A major problem that has been associated with CO₂ removal is solvent degradation and the various problems it presents.

Degradation can lead to solvent loss, corrosion, fouling and foaming. Ionic liquids have low vapor pressure, low regeneration energy and a wide temperature range.

The main disadvantage of ionic liquids is that they are very viscous and expensive (figure 2).

Four ionic liquids (ILs) -tetramethylammonium glycinate (= [Gly]), tetraethylammonium glycinate ([N2222] [Gly]), tetramethylammonium lysinate ([N1111] [Lys]), and tetraethylammonium lysinate ([N2222] [Lys]) were synthesized and mixed with water or N methyl-diethanolamine (MDEA) aqueous solution to form a new type of CO₂ absorption solvents.

The solubility or uptake of CO₂ in these IL + MDEA aqueous solutions was investigated over a wide range of IL concentrations (5–100%), temperature (298–318 K) and CO₂ partial pressure (4–400 kPa).

The results indicated that ionic liquids could greatly enhance the absorption and increased the absorption rate of CO₂ in MDEA aqueous solutions.

It was found that aqueous solutions of 15% IL and 15% MDEA have higher degree of absorption and higher absorption than other 30% IL + MDEA solutions of total amines.

The density and viscosity of ionic liquids are shown in the table below.

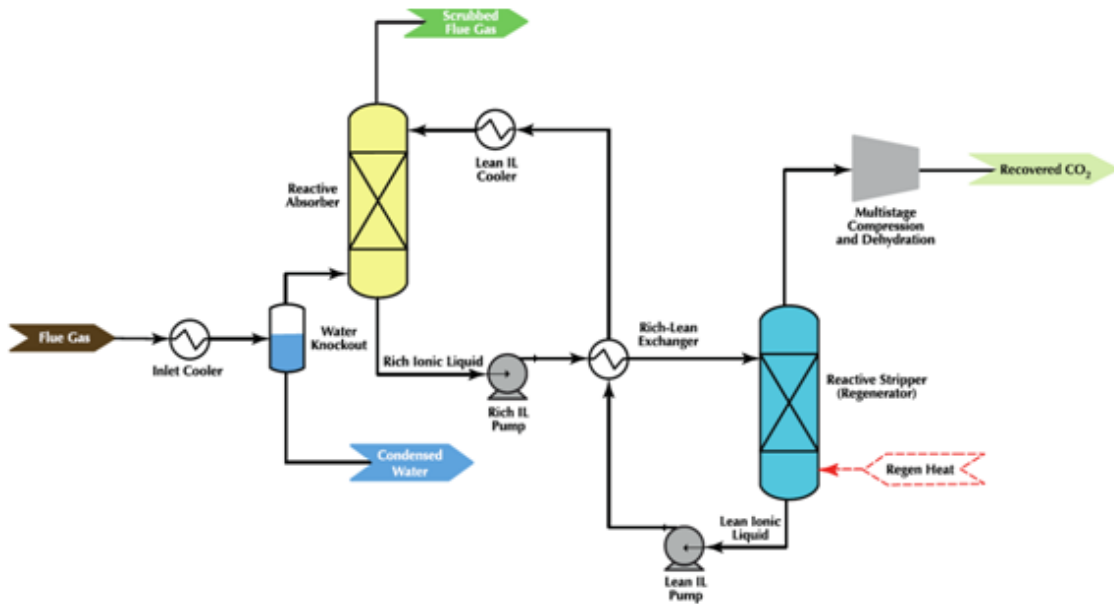
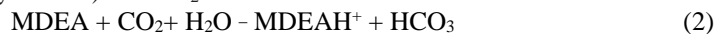


Figure 2. Scheme of the absorption process with ionic liquids[4]

Table 1. Physical parameters of ionic liquids (298 K)[4]

Ionic liquids	Molecular weight	Density ρ (gcm ⁻³)	Viscosity (mPa s)
[N1111][Gly]a	148.18	1.089	304
[N2222][Gly]a	204.35	1.064	129
[N1111][Lys]b	219.11	1.071	569
[N2222][Lys]a	275.28	1.050	672

Mechanism for the reaction of MDEA (tertiary amines) and CO₂:



Since this reaction is essentially a CO₂ hydrolysis under base catalysis, MDEA does not combine with CO₂, absorption is very slow.

For pure amino acid-based ionic liquids, one molecule of CO₂ is combined with two molecules of ionic liquids so the theoretical maximum charge of CO₂ is 0.5 moles per mole of ionic liquids and CO₂ capture from natural gas is accomplished using an n-alkane solvent.

IV. CO₂ SEPARATION IN SUPERSONIC CONDITIONS

In this part of the paper I will deal with the efficient condensation and removal of CO₂ of supersonic separators. A different pressure conditions is analysis of this process[5].

The Laval nozzle created for this experiment and which is a key part of the realization of CO₂ condensation in the supersonic separator has a convergent section according to the Vitosinski curve [5,6], and the construction parameters: $r_0 = 40$ mm, $r_{cr} = 5$ mm and $l_1 = 140$ mm (figure 3).

The convergent section curve are function area by equation[6]:

$$m = m_{cr} = \sqrt{\frac{k}{R} \left(\frac{2}{k+1}\right)^{\frac{k+1}{k-1}} \frac{p^*}{T^*}} A q (Ma) \quad (3)$$

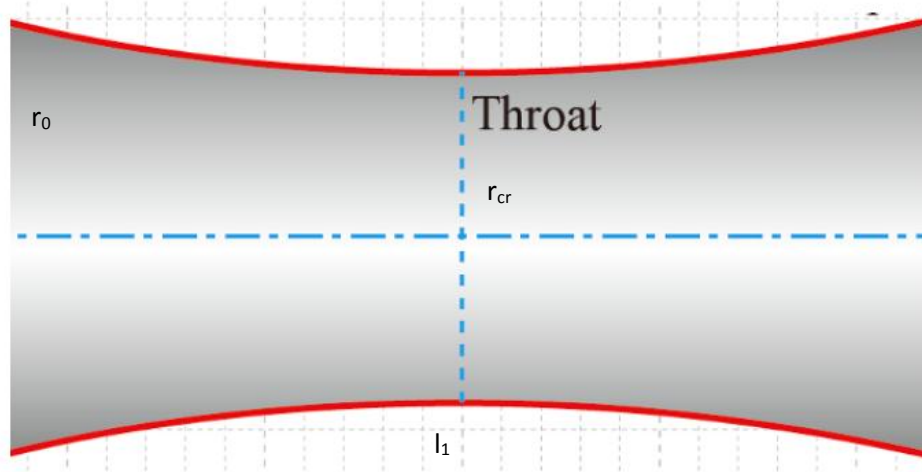


Figure 3.The installation created for the experiment

The governing equations of the gas phase[5,6,7]:

$$\frac{\partial \rho_v}{\partial t} + \frac{\partial(\rho_v u_j)}{\partial x_j} = S_m \quad (4)$$

$$\frac{\partial(\rho_v u_i)}{\partial t} + \frac{\partial(\rho_v u_i u_j)}{\partial x_j} = -\frac{\partial(p_v)}{\partial x_i} + \frac{\partial}{\partial x_j} \left[\mu \left(\frac{\partial u_j}{\partial x_i} + \frac{\partial u_i}{\partial x_j} - \frac{2}{3} \delta_{ij} \frac{\partial u_j}{\partial x_j} \right) \right] + \frac{\partial}{\partial x_j} (-\rho_v \overline{u_i u_j}) + S_u \quad (4.84)$$

$$\frac{\partial(\rho_v E)}{\partial t} + \frac{\partial(\rho_v u_j E + u_j p_v)}{\partial x_j} = \frac{\partial \left(k_{eff} \frac{\partial T}{\partial x_j} + u_i \tau_{eff} \right)}{\partial x_j} + S_h \quad (5)$$

$$S_m = -m_v \quad (6)$$

$$S_u = -m_v u \quad (7)$$

$$S_h = -m_v (h - h_{lv}) \quad (8)$$

The equations governing the flow of the liquid phase are:

$$\frac{\partial \rho(vE)}{\partial t} + \frac{\partial(\rho_v u_j Y)}{\partial x_j} = S_Y \quad (9)$$

$$\frac{\partial \rho(vN)}{\partial t} + \frac{\partial(\rho_v u_j N)}{\partial x_j} = J \quad (10)$$

$$r_d = \left(\frac{3Y}{4\pi \rho_l N} \right)^{1/3} \quad (11)$$

V. DISCUSSION AND CONCLUSION

I ran the above model in Madonna Berkely software, with the gas having an inlet temperature of 273.15 K.

We found that at a temperature of 273.15 K and a CO₂ mole fraction of 20%, the CO₂ condensation position moves forward in the device, at 6 MPa, the condensation start position is at x = 143.27 mm, and at 9 MPa, the position where condensation occurs is at 139.59 mm.

The higher the pressure, the stronger the depression and therefore the greater the nucleation.

After analyzing the variation of nucleation rate and droplet radius (Fig. 6 and 7), it can be concluded that, the maximum nucleation rate of CO₂ gradually decreases and the radius of CO₂ droplets gradually increases.

This is because the higher the inlet pressure, the earlier condensation occurs, the higher the temperature at which condensation occurs, and the lower the nucleation rate.

After droplet formation, CO₂ removal is achieved only if the partition wall captures the liquid film and allows it to flow to the collector.

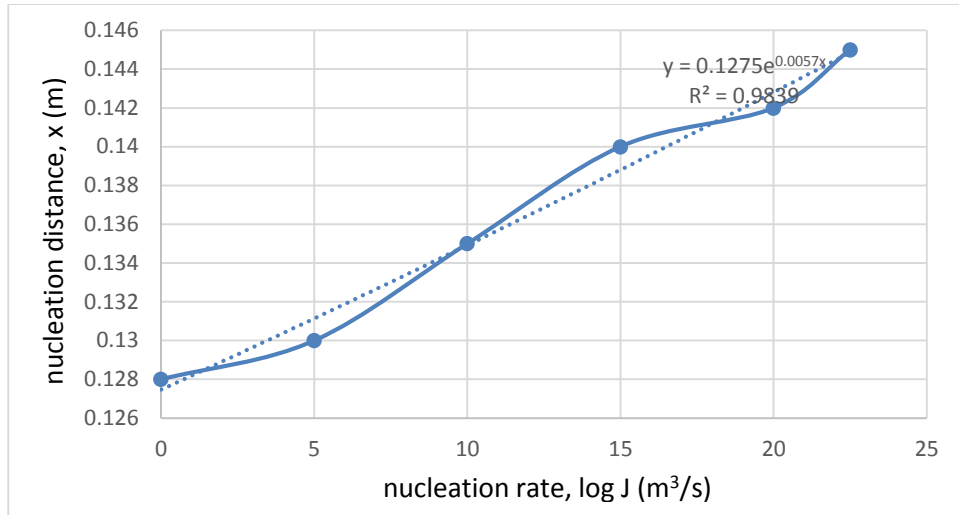


Figure 4. Nucleation distance (y (m)) versus spontaneous nucleation rate x (log J (m³/s)) at 6 MPa pressure

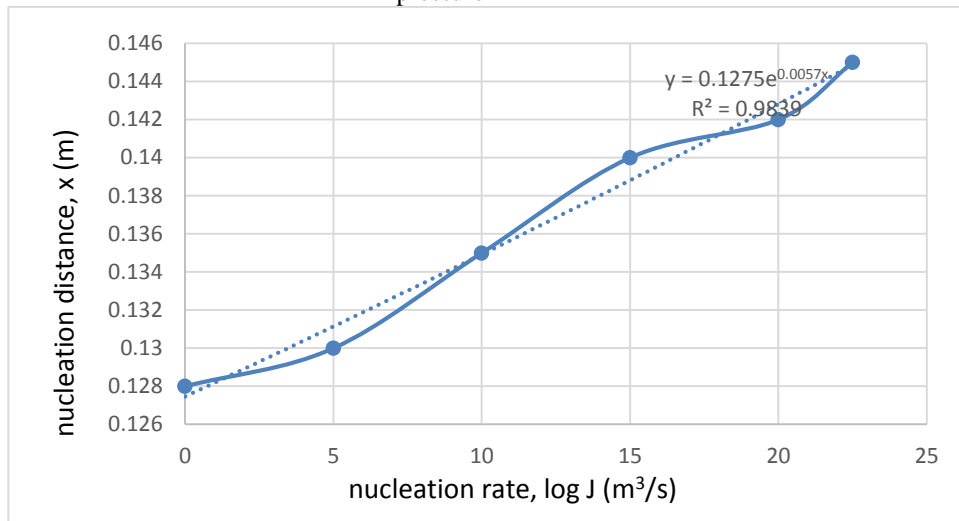


Figure 5. Nucleation distance (y (m)) versus spontaneous nucleation rate x (log J (m³/s)) at 7 MPa pressure

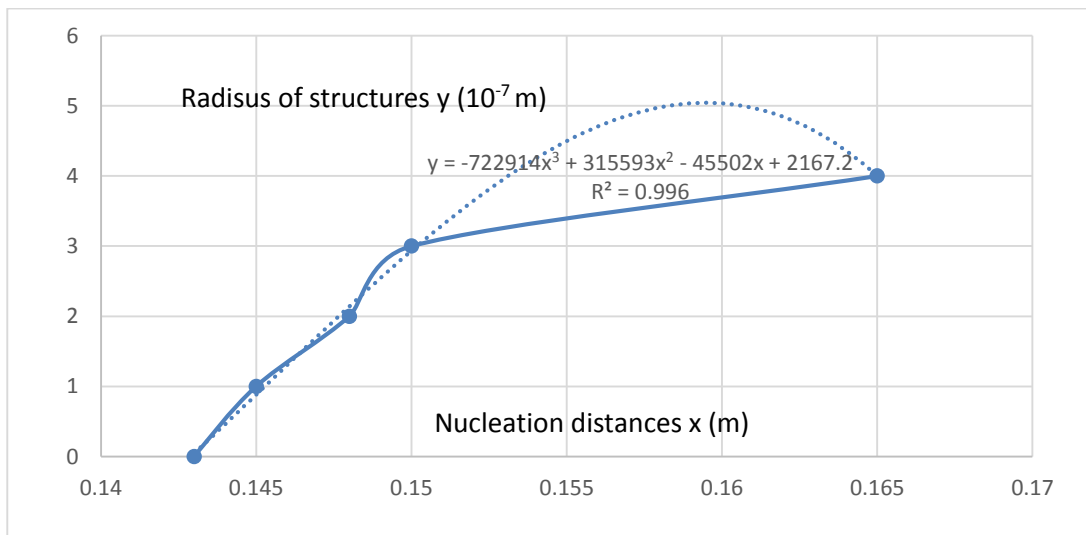


Figure 6. Radius of the drop (cluster) y (10⁻⁷ m) as a function of the nucleation distance x (m) at the pressure of 6 MPa

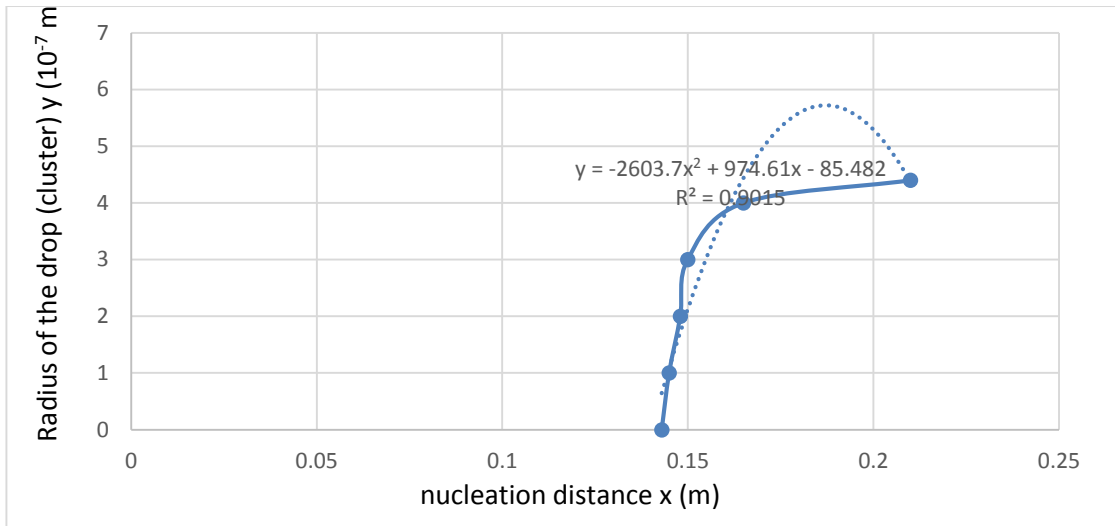


Figure 7.Radius of the drop (cluster) y (10^{-7} m) as a function of the nucleation distance x (m) at the pressure of 7 MPa

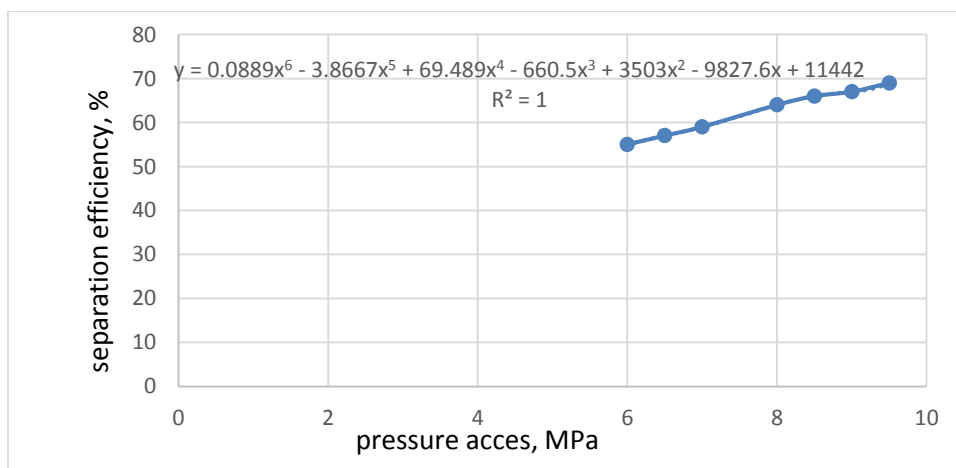
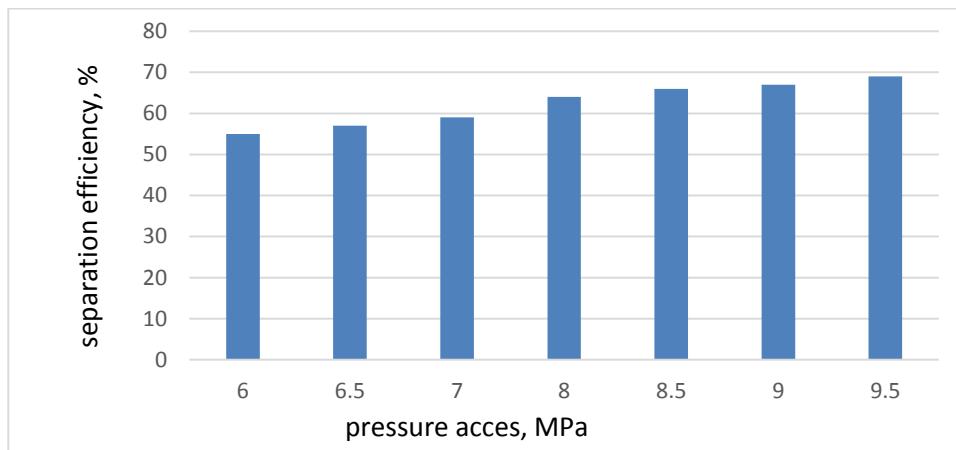


Figure 8.Separation efficiency y (%) as a function of inlet pressure x (MPa)

Therefore, in addition to the liquefaction efficiency, the separation efficiency is also an important index to evaluate.

During the separation process, some of the liquefied CO₂ droplets enter the collection tank. Instead, the remaining part flows from the dry gas and is then regasified.

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