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Ammonia absorption: Implications of the absorber type

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The aim of this work is to carry out a comparative study between the falling film absorber, trays and packed absorber columns applied to the absorption refrigeration system using the mixture of ammonia and water as the working fluid. For this comparison, the concentration of ammonia in the liquid phase at the outlet of the absorbers was used as the design variable. In order to carry out this study, simulations of the tray and packed absorber columns were carried out using the Aspen Plus commercial simulator. The simulations of the falling film absorber were carried out in Matlab, as Aspen Plus does not have a model for this equipment in its operating packages. The results of the simulations show that the falling film absorber is the most suitable for the process in question, as it needs smaller dimensions to reach the design concentration. The comparative study between the columns shows that the packed column requires a smaller number of stages and a lower height to reach the design concentration. As such, this study motivates future work, especially regarding an economic evaluation of the absorbers investigated here.
Keywords: *refrigeration cycle, falling film, filling column and column*

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I. Introduction

The most commonly used absorbers to ammonia absorption are the falling film andpacked columns, while trays columns can also be used to a lesser extent. With regard to falling film absorbers, the designs presented in the literature take into account the type of device (plates, vertical tubes and horizon tubes) used to form the film and their respective quantities and dimensions. Goel and Goswamy (2005a), for example, presented a falling film absorber design using Alloy 326 plates. In the configuration proposed by the authors, the steam and cooling fluid (water) flow from the bottom to the top of the absorber, while the liquid solution of ammonia and water flows from the top to the bottom. The authors observed good performance from this absorber for the system in question.Goel and Goswamy (2005b) propose a new design for falling film absorbers made up of horizontal tubes. They evaluate the influence of increasing the number of cooling fluid tubes on ammonia absorption rates. To do this, they reduced the diameter of the tubes and compared them to a horizontal absorber with fewer tubes and a larger diameter.The authors concluded that this new design reduces the size of the equipment by 25% and increases the mass transfer area. This results in more efficient and compact equipment. Triché et al. (2016) presented a prototype plate-type absorber in which the vapor and liquid solution flow concurrently from the top to the bottom of the absorber, while the cooling fluid flows countercurrently from the bottom to the top.The authors found good absorption performance with a 26% increase in the concentration of ammonia in the liquid phase. The design proposed by Aminaravy et al. (2017) consists of a falling film absorber made up of vertical tubes. The tubes are arranged in a column where the cooling fluid (water) flows from the top to the bottom of the column, while the vapor and the liquid solution of ammonia and water flow from the top to the bottom. With this configuration, the authors found that the absorption system performed well.

With regard to the application of packing columns for the absorption of ammonia by a solution of ammonia and water, Selim and Elsayed (1999a) investigated Rasching rings and ceramic Berl saddles as types of packing. Based on the results, the authors concluded that ceramic saddles perform better compared to Rasching rings. In addition, they suggest that the process be carried out with continuous cooling of the solution, given that absorption is exothermic. As a continuation of their previous work, Selim and Elsayed (1999b) evaluated the performance of this type of absorber in absorption refrigeration cycles. The authors presented a mathematical model to represent the equipment and carried out a performance study under various design and operating conditions. As in Selim and Elsayed (1999a), two types of filling were investigated: Rasching rings and ceramic Berl saddles. The authors concluded that when ceramic Berl saddles are used, a lower filling height is required. In addition, the results of this work show that the absorption process is affected by the following parameters: cooling fluid flow rate, bed height, steam and solution flow rates and the inlet conditions and filling type.The authors also found that changing the pressure of the bed and/or the temperature of the steam inlet did not influence the performance of the equipment. Fernández-Seara et al. (2002) proposed a model to represent a packed column in a refrigeration system with ammonia and water absorption. The filling used by the authors was ceramic Berl saddles.With this type of packed and under the absorber's operating conditions, the authors obtained good performance in the operation of this equipment. Sieres and Fernández-Seara (2007) developed a model to simulate the simultaneous heat and mass transfer processes that occur in the main components of a heat absorption systemNH_3-H_2O. The model was applied and validated with data from an absorption column with three different types of fillings: ceramic Berl cells, raschig rings and Novalox. The results of the research showed that ceramic Berl cells performed better. This type of filling was then used to carry out various simulations of the equipment in question. To the best of the authors' knowledge, applications of plate-column absorbers for this system have been studied in the literature no less.

The need to increase heat and mass transfer rates, combined with the desire for increasingly compact and efficient components, has led researchers to idealize, model and experimentally evaluate various absorber configurations applied to the ammonia and water absorption refrigeration cycle.Given the possibility of using different equipment to absorb ammonia from a solution of ammonia and water, a study of the efficiency of this equipment makes a significant contribution to the literature and the industry, as the absorber is crucial to the good performance of the absorption refrigeration cycle. Each absorber has its own peculiarities, favorable and unfavorable aspects for the absorption process, as can be seen in Table 1. In this context, the aim of this work is, through simulations in Matlab and Aspen Plus, to carry out a comparative study between the ammonia absorption capacities of absorbers: falling film plate, packed column and plate column. It is important to note that although the literature proposes different designs for absorbers, there is a lack of comparative studies between them. Aspects such as the length of the equipment, the diameter of the columns of plates and filling, the type of filling, the type of plates, and the influence of the inclusion of the cooling system in the absorber columns are explored in this work. In addition, it should be noted that the literature lacks an analysis of the trays absorber column applied to ammonia absorption, so this application is another contribution of this article.

Table1-Characteristics of falling film, packed column and plate column absorbers.

PARAMETERS	TYPE OF ABSORBER		
	FallingFilm	**PackedColumn**	**TraysColumn**
Mass Transfer	Lowmasstransfer rates.	Mass transferis high.	The mass transfer rates are the highest.
Interfacial Area	The interfacial area between the vapor and the liquid is low.	The packed increases the interfacial area between the liquid and the vapor.	A large volume of vapor comes into contact with the liquid solution. Therefore, a larger interfacial area.
Heat transfer and heat removal	The liquid solution cooling system is easily incorporated into the equipment.	Incorporating the liquid solution cooling system is a difficult task.	Incorporating the liquid solution cooling system is simpler than in packed columns.
Pressuredrop	Low pressure drop compared to the column of trays and packed	Low pressure drop compared to plate columns.	High pressuredrop

The article is organized as follows: first, in section 2, the methodological aspects of the absorber simulations are presented. Section 3 presents the performance results of each absorber and discusses the comparative study. Finally, thepaperconcludes.

II. Methodologicalaspects

The purpose of this section is to present the methodological aspects used for the comparative study of the absorbers: falling film, packed column and trays column. To make it easier to present the results, this paper will use the following nomenclature for the absorbers:

- ABS 1- Falling film absorber
- ABS 2- PackedColumn
- ABS 3- TraysColumn

Therefore, firstly, aspects related to thermodynamic models to describe the ELV of the ammonia and water system are presented. This is followed by a discussion of the parameters that must be taken into account when designing the absorbers studied in this work, as well as the design variable to be considered. Subsequently, how the simulations will be processed, information such as software and model simplifications are discussed. Finally, the operating conditions for the absorbers used in the simulations are presented.

a. Selection of thermodynamic models to describe the ELV of the ammonia and water system

For the description of the liquid-vapor equilibrium of the ammonia-water mixture, the literature(Goel e Goswami, 2005a, 2005b; Sieres e Fernández-Seara, 2007; Triché*et al*, 2016, 2017; Aminyavari*et al.*, 2017) has used empirical correlations proposed by Pretek and Klomfar (1995). However, these correlations leave the model inflexible, since the correlations are not valid for other working fluids. In view of this, Rosa et al. (2020) successfully proposed rigorous thermodynamic modeling, as presented in Prausnitiz et al. (1978), todescribetheliquid-vapor equilibrium in question.In this approach, the vapor phase is considered an ideal gas and the non-idealities of the liquid phase are described by a hyperbolic tangent function proposed by Prausnitiz et al. (1978). Further details of this thermodynamic model and its validation can be found in Rosa et al. (2020) and Prausnitiz et al. (1978). Therefore, this more rigorous approach will be used in this work to calculate the liquid vapor balance of ammonia and water in the falling film absorber. However, this approach cannot be used in the simulations of the absorber columns because Aspen Plus, although it has a wide variety of thermodynamic models in its computational packages, does not have the model proposed by Prausnitiz et al. (1978).

The UNIQUAC (UNIversalQUAseChemical) model proposed by Abrams and Prausnitz (1975) was chosen to describe the non-idealities of the phase, as recommended by Kondrashina (2018) and Rosa et al. (2021).The UNIQUAC model is based on the concept of local composition and states that the composition of the system in the vicinity of a given molecule is not the same as the composition of the mixture; this is due to the difference in size and interaction energies of the molecule. For the vapor phase, ideal gas was considered, as recommended by Rosa et al. (2020) and Triché et al. (2016) for the pressure in question (6 bar). Therefore, in order to verify the predictive capacity of the UNIQUAC model to describe the non-idealities of the liquid phase combined with the ideal gas consideration for the vapor phase, liquid-vapor equilibrium simulations were carried out for the ammonia-water mixture at 90°C. The simulations were validated with data from the literature.

2.2 Absorber designs: falling film, packed column and trayscolumn

This work, using simulations, will carry out a comparative study between absorbers: falling film, trays and packedcolumn. The study will evaluate the design conditions of the absorbers needed to achieve a certain concentration of ammonia at the outlet of the liquid solution, since relatively high ammonia concentrations reflect higher absorption rates. Therefore, the expected concentration at the outlet of the absorbers must first be defined. To this end, a literature review was carried out on the types of absorbers and variations in the concentration of the liquid stream, as shown in Table 2.

Table2-Concentration at the liquid outlet of NH_3-H_2O absorbers

Reference	Absorver	$x_{NH_3}, inlet$	$x_{NH_3}, outlet$
Goel e Goswamy (2005a)	*Falling film*	0,25	0,37
Goel e Goswamy (2005b)	*Falling film*	0,25	0,35
Sieresand Fernández-Seara (2007a)	*PackedColumn*	0,24	0,31
Triche*et al.*(2016)	*Falling film*	0,46	0,58
Triche*et al.*(2017)	*Falling film*	0,46	0,58

Table 2 shows that the most significant increases in concentration at the outlet of the liquid stream are observed in the falling film type absorbers proposed by Triche et al. (2016) and Triche et al. (2017), with an increase of around 26% in relation to the feed concentration. Therefore, the concentration of will be used in this work as the parameter to be achieved in the falling film, trays column and packed column absorbers. It is important to note that, to the authors' knowledge, the literature does not report optimum ammonia concentration values at the outlet of the liquid stream. Therefore, the following are aspects related to the design of the absorbers to be considered in this work. The height and stages of the columns and the length of the falling film will not be discussed in this section as they are the subject of study in the results section of this work.

For the falling film absorber, the prototype proposed by Triche et al. (2016) was taken as a reference. This is a plate-type absorber in which the vapor and liquid solution flow concurrently from the top to the bottom of the absorber, while the cooling fluid flows countercurrently from the bottom to the top. The absorber studied consists of 16 plates: 8 channels for the cooling fluid and 7 channels for the liquid solution and vapor. There are therefore 14 falling films formed between the plates. Table 2 shows the geometric dimensions of the falling film from Trciheet al.(2016).

Tabela 3-GeometricdimensionsfromTrciheet *et al.*(2016).

Typeofplate material	Liga de metais 316 (Alloy 326)
Plate width	111 mm
Plate thickness	0,4 mm
Numberofplates	16

According to Strigle (1987), the design procedure for an absorption column, be it a plate or a packed column, requires the determination of the column's diameter and height. Therefore, for the absorber columns studied in this work, these two parameters will be evaluated. If the inside of the absorber column is filled, the literature has used raschig rings, Novalox and ceramic Berl cells for the system in question. Although Selim and Elsayed (1999a) report that when ceramic saddles are used instead of Rasching rings, the absorption rate increases by 5% to 8% for a given solution and steam flow rate, this work will carry out a comparative study between these two types of packed.With regard to the diameter of filling column absorbers, Treybal (1955) and Chavez-Islas and Heard (2009) state that if the diameter of random packed columns, as is the case here, is greater than 0.60 meters, the distribution of liquid inside the column may be compromised. Therefore, the study of the influence of the diameter of the packed column on ammonia absorption will be carried out taking this into account.

If the absorber has internal trays, Treybal (1955) recommends that the plates should be of the movable valved type for absorbers. However, the literature does not show the performance of this equipment with another type of trays for the system in question. Therefore, in this work, valved and perforated plates will be studied for ammonia absorption. With regard to the diameter of the plate column, Perry and Chilton (1999) state that plate column absorbers are preferred for diameters greater than 0.60 meters; for smaller columns, it is recommended to use packed as internals. Therefore, this value will be taken into account in the range of diameters studied for column absorption.

In addition, according to Selim and Elsayed (1999a), a continuous cooling system for the solution is necessary since the absorption of gaseous ammonia by a solution of ammonia and water is exothermic. Therefore, the cooling system is essential to keep the solution away from the boiling point and to maintain the temperature required for absorption in all the absorbers studied in this work. Therefore, as with the absorbers studied in the literature (Krishnamurth and Taylor, 1986, Selim and Elsayed, 1999a, Elsayed, 1999b, Kang et al., 2000, Fernández-Seara et al., 2002, Fernández-Seara et al, 2002, Goel and Goswamy, 2005a, Goel and Goswamy, 2005b, Sieres and Fernández-Seara, 2006, Bohra, 2007, Sieres and Fernández-Seara, 2007, Chavez-Islas and Heard, 2009, Triché et al., 2016 and Aminaravy et al., 2017) for the comparative study presented here, the cooling system integrated into the three absorbers will be taken into account. In addition, this study will evaluate the impact of not considering the cooling system in the plate and filling absorption columns. In view of the above, Table 4 presents a summary of the design characteristics of the absorber columns that will be studied in this work.

Table 4- Design characteristics of trays and packed absorbers

ABS 2: TraysColumn	**ABS 3**: PackedColumn
Valved plates and perforated plates Column diameter: minimum 0.60 m With and without cooling system	Filling type: Rasching rings Column diameter: maximum 0.60 m With and without cooling system

b. **Aspectsrelatedto absorber simulations**

The simulations of the absorbers: trays column and packed are carried out in Aspen Plus. This is one of the most widely accepted commercial simulators on the market and makes it possible to study the stationary and dynamic simulation of various industrial equipment, as well as having a variety of thermodynamic models to describe the phase equilibrium in the various systems to be simulated. Using the RadFrac computational package, Aspen Plus allows you to simulate absorption columns and add elements to them, such as tryasand packings.Therefore, in order to simulate the absorber columns, the system (ammonia and water) must first be selected, as well as the thermodynamic models to describe the liquid-vapor equilibrium. The UNIQUAC thermodynamic model will be used to describe the non-idealities of the liquid phase and the vapor phase will be considered as an ideal gas, as explained in the previous section. Subsequently, in the RadFrac tool options, the absorber column to be simulated must be chosen. Once this is done, the equipment's inlet and outlet streams are added, as well as information on the inlet, such as temperature, pressure and composition. For the case study here, the liquid stream is fed at the top of the column while the gaseous stream is fed at the bottom. In addition, the cooling system must be added. This is a liquid water stream fed into the base of the column; inlet conditions, such as temperature and pressure, of this stream must be specified.Once the input currents have been determined, the elements must be added to the absorber column along with their specifications. For a trays absorber column, information such as: plate type, column diameter, number of stages and column height must be specified. For a packed column, the following must be established: the type of packed and its dimensions, the number of stages and the height of the packed or the equivalent height per theoretical plate (HEPT). Once the column specifications have been established, simulations can be carried out. It is important to note that, despite the potential of this simulator, Aspen Plus does not have any models that describe the behavior of a falling film absorber.

The model proposed by Rosa et al. (2020) was used to describe the falling film absorber. This is described by differential mass and energy balances, together with the algebraic equations for mass and energy transfer and the phase equilibrium correlations at the interface. Thermodynamic and transport properties are calculated via the CAPE-OPEN interfaces using the libraries in TEA (Thermodynamics for Engineering Applications). To calculate the phase equilibrium at the interface, Rosa et al. (2010), contrary to the empirical correlations used in the literature, used the model proposed by Prausnitiz et al. (1978) to calculate the non-idealities of the liquid phase and the vapor phase was considered an ideal gas. Therefore, greater flexibility and thermodynamic formalism are added to the model.

The resulting set of equations consists of a system of non-linear differential algebraic equations. The algebraic equations were discretized using the finite difference method, resulting in an algebraic system. This was implemented in MOSAICmodeling (http://mosaic-modeling.de/), which is a free web-based modeling environment capable of automatically generating code for process simulation and optimization in different programming languages (Merchan et al., 2015 and Tolksdorf et al., 2019). The model is solved in Matlab using the fsolve function, which is based on the residual minimization method.

c. OperationalConditions

Once the physical characteristics of the absorbers have been defined, as well as the design parameter (mole fraction at the absorber outlet), the operating conditions are established in this subsection. The comparative study of absorber efficiency was carried out for a nominal case, where the input operating conditions are shown in Table 5. These conditions are the same as those adopted in the falling film absorber presented in the work by Triché et al. (2016).

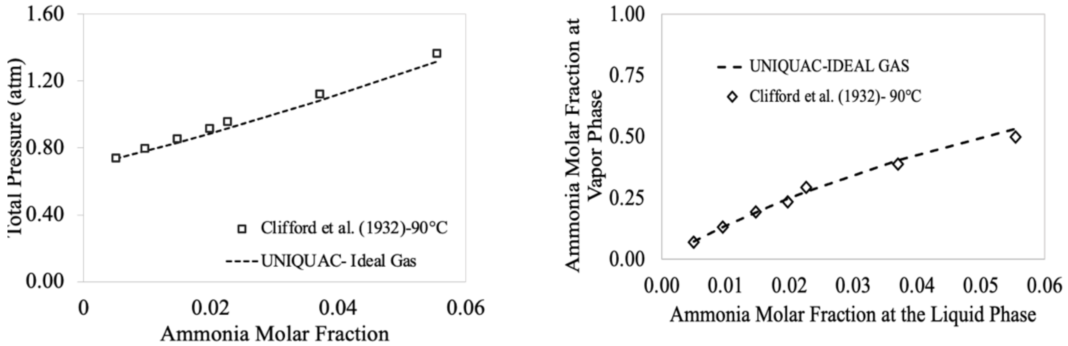
Table5- Operating conditions Ammonia absorption at 6 bar pressure.

Variable	Vapor Inlet	LiquidInlet
Mass Flow-rate	(kg/s)	(kg/s)
Temperature	(K)	312.0 (K)
NH3 molar fraction	0.995	0.46

III. Results and discussion

AThe absorber simulations were carried out for a nominal case shown in Table 4 and the design specifications were as reported in Table 2 for the fallinf film absorber and Table 3 for the plate and filling columns.

The results of the simulations are shown in Figure 1 and were compared with experimental data from the literature. The UNIQUAC model combined with the ideal gas was found to accurately describe the experimental total pressure data (Figure 1), with average deviations of 3.46%. Good accuracy was also observed in the reproduction of the vapor phase mole fraction, as shown in Figure 1, with an average deviation of 4.48%.



(a) (b)

Figure 1- Validation of the UNIQUAC thermodynamic model for the ELV of the NH_3-H_2O system (a) Total pressure data and (b) ammonia mole fraction data in the vapor phase

In order to evaluate the amount of ammonia absorbed, Figure 2a shows the variation in the amount of ammonia absorbed in the liquid phase in relation to the height of the equipment for the absorbers: ABS 1, ABS 2 and ABS 3. Figure 2b shows the temperature behavior in relation to the lengths of the absorbers. Figure 3 studies these variables in relation to the number of stages with and without the integrated cooling system for the trays column and the packed column. In Figures 2 and 3, the horizontal red line represents the design variable (amount of ammonia absorbed by the liquid phase).

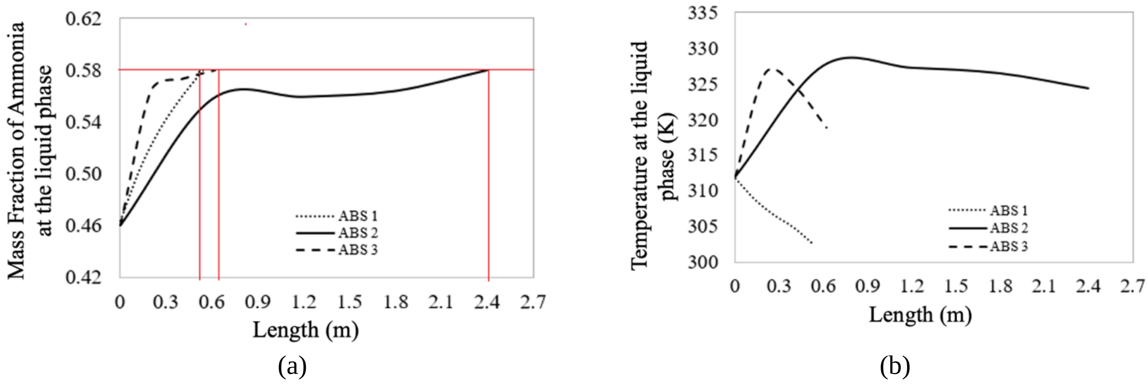


Figure 2- Variation of (a) amount of ammonia absorbed in the liquid phase and (b) temperature in relation to the height of the equipment for the absorbers: ABS 1, ABS 2 and ABS 3.

Looking at Figure 2a, it can be seen that thetray column absorber requires a greater height (2.4 meters) to reach the design concentration (0,58), while the falling-film absorber reaches it with a length of 0.526 meters and the packed column at 0.620 meters. It is therefore clear that, for the conditions evaluated here, the falling film absorber is the most compact piece of equipment for the process in question. Kang et al. (2000) and Oliva et al. (2009) also point out that the falling film absorber has some advantages over the others, such as: the same interfacial heat transfer mass area, compact and operationally stable equipment. The packed column also proved to be a good alternative for ammonia absorption, while the plate column requires a height approximately 4 times greater than that of the other equipment.In addition, Figure 2b shows that the temperature of the falling film decreases along the absorber, while in the columns there is an increase at the beginning of the absorber and then a drop. This drop is less marked in the trays column. This shows that it is difficult to control the temperature in the columns studied here. As stated by Govindaraju (2005), incorporating a cooling system into absorber columns is a more difficult task compared to falling film.

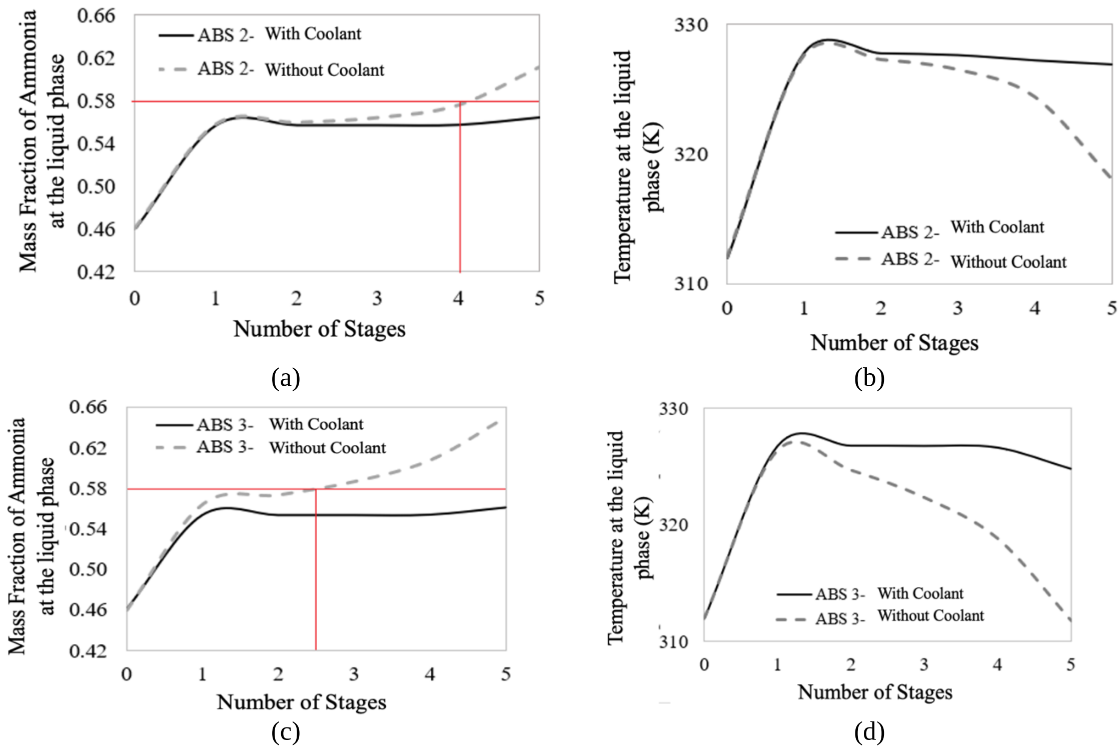


Figure 3- Variation of the mass fraction of ammonia absorbed in the liquid outlet and temperature with number of stages with and without the presence of the cooling system for (a) and (b) tray column and (c) and (d) packed column

Looking at Figures 2a, 3a and 3c, it can be seen that, as expected, the concentration of ammonia increases along the length of the absorbers, which indicates that ammonia is continuously absorbed by the liquid phase. In addition, it can be seen that the absorption of ammonia in the liquid phase occurs more significantly at the beginning of the absorbers. This may be due to the fact that the concentration of ammonia in the liquid phase is lower, so the concentration gradient (driving force for mass transfer) is greater, which favors the absorption of ammonia. This considerable absorption of ammonia at the beginning of the absorber is reflected in the significant increase in the temperature of the liquid stream in absorbers ABS 2 and ABS 3, as can be seen in Figures 2b, 3b and 3d. This behavior is associated with the exothermic absorption of ammonia; higher absorption rates imply a greater amount of heat transferred to the liquid phase, so an increase in temperature is observed. This behavior is not seen in the falling film absorber (Figure 2b), showing that the falling film cooling system is more efficient than the others.

As shown in Figures 3a and 3c, it can be seen that thepacked absorber, ABS 3, needs 3 stages to reach the ammonia design concentration of 0.58, while the prtayscolumn, ABS 2, needs 4 stages. This shows that, compared to the trays column, the packed column is more suitable for the process in question. It is more compact and can reach higher concentrations of ammonia in the liquid phase. Furthermore, the addition of a cooling system to the columns is essential for efficient operation. Without it, the design concentration would not be reached, or a very large number of stages would be needed to achieve it, making their application unfeasible.

IV. Conclusion

This article presents a comparative study of three types of absorbers (falling film, trays column and packed column) for ammonia absorption. The simulations carried out aimed to specify the physical characteristics of the absorbers for the design variable: concentration of ammonia in the liquid phase at the outlet of the absorbers. The ammonia concentration profile and the temperature profile in the liquid phase were studied in the three configurations. In all the scenarios investigated, it was found that the most significant ammonia absorption occurs at the beginning of the columns. For the system studied, under the specific operating conditions, the falling film absorber is more efficient than the others, as the design concentration was reached in shorter lengths. It also has a more compact arrangement than the others. This makes it possible to integrate it with other equipment, such as pre-absorbers, by intensifying processes, as well as making it more versatile in the sense that it can be applied in industries where physical space is limited, such as offshore platforms. When comparing the columns, it can be seen that thepacked column shows better results than the trays column, which is a larger piece of equipment. These results motivate future studies, especially with regard to a comparative economic analysis of the absorbers.

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