MODELING AND SIMULATION OF HMT ON NATURAL GAS DEHYDRATION USING SOLID DESICCANTS

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Abstract: Natural gas is the fastest growing primary energy source and its use is expected to almost double to 2020, providing a relatively more clean fuel for the worldwide energy demand. Some contaminants, such as water vapor, have to be removed before its use. Dehydration of natural gas is of extreme importance to avoid the formation of hydrates and other associated problems. Different dehydration methods exist; however the solid desiccant approach can dehydrate natural gas to levels of water vapor lower than other methods. This paper presents a simple formulation for simulating the dehydration of a wet methane stream flowing though a porous medium composed of hygroscopic particles. The model assumes small resistance to diffusion within particles and takes heat and mass transfer rates into account by employing constant transfer coefficients. The formulation was developed based in the traditional mass and energy conservation principles, considering a one-dimensional transient scenario. A dimensional analysis is also performed and relevant dimensionless groups are introduced. The set of dimensionless parameters are similar to effectiveness-NTU groupings, widely used in heat exchangers. Despite its simplicity, the proposed formulation presents a new contribution since similar analysis is apparently unavailable in the gas dehydration literature. Finally, in order to illustrate the effect of varying the dimensionless groups the formulation is solved using a combined finite-volumes/method-of-lines approach. The results example how the dimensionless parameters can influence the dehydration process.

Keywords: Natural Gas, Adsorption, Dehydration, Solid Desiccant, Dehumidification.

1. NOMENCLATURE

\mathcal{A}	area	Subsc	ıbscripts	
c_p	constant pressure specific heat	adv	advective	
h_h	convective heat transfer coefficient	b	bed	
h_m	convective mass transfer coefficient	δ	closed pore	
i	specific enthalpy	e	effective or apparent	
j''	mass flux	g	natural gas	
L	cylinder height	in	inlet	
m	mass	i	process gas flow	
\dot{m}	mass flow rate	l	adsorbed water (liquid phase)	
t	time	\max	maximum	
T	temperature	\min	minimum	
v_i	process gas flow velocity	op	operation	
v_b	bulk velocity	out	exit	
x	distance from the cylinder entrance	p	particle	
W	dry basis adsorbed water concentration	π	pore	
Y	dry basis vapor concentration	s	solid phase or surface	
Greek Symbols		v	water vapor	
ρ	density	Super	Superscripts	
ϵ	total porosity	*	dimensionless quantity	
au	periodical time	\sim	dry basis	

2. INTRODUCTION

From the standpoint of processing and conditioning of natural gas, it should be brought to water levels established by oil and natural gas standards in order to prevent the formation and deposition of solid hydrates (Carroll, 2009) that may in the long run reduce the useful diameter of pipes (Jassim *et al.*, 2010; Mokhatab *et al.*, 2007). Furthermore, it can block partly or wholly valves, corrosion formation (Obanijesu *et al.*, 2011) and reduce equipments life expectancy.

To prevent hydrate formation, different techniques may be used. Among them, there are processes more widespread such as liquid and solid desiccant for gas dehumidification.

Work made by Gandhidasan *et al.* (2001) performs a study on the use of solid desiccants based on silica gel regarding the effects in various parameters changes. Moreover, a study was developed on the efficiency, i.e. to estimate the energy required in the regeneration process. Recently, Nastaj and Ambrozek (2009) performed a theoretical analysis of the adsorptive drying process of gases in a cyclic temperature swing adsorption system with two fixed bed adsorption columns using silica gel as solid desiccant. The computer simulations results are used to study the effects of the inlet gas relative humidity on the breakthrough curves in the adsorption step and purge gas temperature on the breakthrough curves in the desorption step.

Apparently, a notable feature of previous works on dehydration of natural gas using solid desiccant is a few development compared to other methods of natural gas dehydration. In this context, this work presents a mathematical formulation for heat and mass transfer for desiccant dehydration of natural gas. The formulation herein presented is based on normalized dimensionless groups used in studies like those developed for heat and mass exchangers (Sphaier and Worek, 2009) and for storage of adsorbed natural gas (da Silva and Sphaier, 2010).

3. PROBLEM FORMULATION

In this section, the main formulations for the dehydration of natural gas using solid adsorbents are presented. The reservoir has only one entrance and one exit for the natural gas flow. Initially, the reservoir is empty and the pressure and temperature are equal to the surroundings. Then, as the natural gas with higher levels of water vapor flows into the reservoir the process of dehumidification begins.

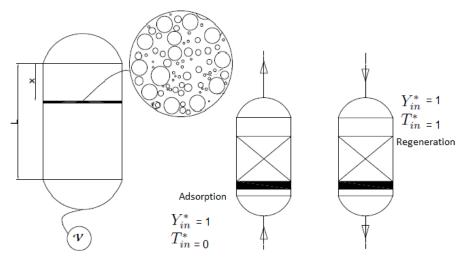


Figure 1. Volume, porous area view and dehydration cycle configuration

According to figure 1, two steps are done over dehydration cycle: adsorption and regeneration. The former intend to reduce the natural gas water vapor concentration and the latter is required to remove water of the solid desiccant providing the possibility of restarting the cycle.

The simplifying assumptions considered in the derivation of the current model are described such as: natural gas as water vapor and methane; incompressible flow; negligible pressure drop or loss; unidirectional flow; constant specific

heats; radiation effects are negligible due to the relatively low temperature differences; there is no chemical reaction nor any internal energy generation of any form; isotropic medium; no phase change besides that resulting from adsorption; and no heat losses to the environment.

3.1 Mass Balance

Since the mass of the solid portion and that of the natural gas are assumed invariable only a mass balance for water is required. Denoting \mathcal{V} as a portion of the vessel volume (that includes all volumes) and \mathcal{S} as its bounding surface, an integral mass balance is written as:

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{x_0}^{x_1} \left(\epsilon_b \,\rho_{v,i} + (1-\epsilon_b) \,\epsilon_p \,\rho_{v,\pi} + \rho_l\right) \mathcal{A}_x \,\mathrm{d}x = -\left(\boldsymbol{j}_{v,i}^{\prime\prime} \cdot \hat{\boldsymbol{n}} \,\mathcal{A}_x\right)_{x=x_1} + \left(\boldsymbol{j}_{v,i}^{\prime\prime} \cdot \hat{\boldsymbol{n}} \,\mathcal{A}_x\right)_{x=x_0} \tag{1}$$

where $\mathbf{j}_{v,i}'' = \rho_{v,i} \mathbf{v}_b = \epsilon_b \rho_{v,i} \mathbf{v}_i = \rho_g Y \mathbf{v}_b$ and $S = \mathcal{A}_s / \mathcal{V}$ Finally, simplification yields:

$$\epsilon_b \frac{\partial Y}{\partial t} + (1 - \epsilon_b) \epsilon_p \frac{\partial Y_p}{\partial t} + \frac{\rho_b}{\rho_g} \frac{\partial W}{\partial t} + v_b \frac{\partial Y}{\partial x} = 0$$
⁽²⁾

A general mass balance for the particles volume is given by:

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{x_0}^{x_1} \left(\rho_l + (1 - \epsilon_b) \epsilon_p \,\rho_{v,\pi}\right) \mathcal{A}_x \,\mathrm{d}x = \int_{x_0}^{x_1} h_m \,\rho_g \left(Y - Y_p\right) \mathcal{S} \,\mathcal{A}_x \,\mathrm{d}x \tag{3}$$

After simplifications, one can conclude that:

$$\frac{\rho_b}{\rho_g} \frac{\partial W}{\partial t} + (1 - \epsilon_b) \epsilon_p \frac{\partial Y_p}{\partial t} = h_m \mathcal{S} \left(Y - Y_p \right) \tag{4}$$

Subtracting equation (4) from (2) yields a mass balance for the process gas flow:

$$\epsilon_b \frac{\partial Y}{\partial t} + v_b \frac{\partial Y}{\partial x} = -h_m \mathcal{S} \left(Y - Y_p \right) \tag{5}$$

3.2 Energy Balance

Since pressure variations are assumed negligible, the following relations apply:

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{x_0}^{x_1} \left(\epsilon_b \,\rho_g \left(i_g + i_{v,i} \,Y\right) + \left(1 - \epsilon_b\right) \epsilon_p \,\rho_g \left(i_g + i_{v,\pi} \,Y_p\right) + i_l \,\rho_b \,W + i_s \,\rho_b\right) \mathcal{A}_x \,\mathrm{d}x = \\ = -\left(\rho_g \left(i_{v,i} \,Y + i_g\right) \left(\boldsymbol{v}_b \cdot \hat{\boldsymbol{n}}\right) \mathcal{A}_x\right)_{x=x_1} + \left(\rho_g \left(i_{v,i} \,Y + i_g\right) \left(\boldsymbol{v}_b \cdot \hat{\boldsymbol{n}}\right) \mathcal{A}_x\right)_{x=x_0} \quad (6)$$

Finally, simplification yields:

$$\epsilon_b \frac{\partial}{\partial t} (i_g + i_{v,i} Y) + (1 - \epsilon_b) \epsilon_p \frac{\partial}{\partial t} (i_g + i_{v,\pi} Y_p) + \frac{\rho_b}{\rho_g} \frac{\partial}{\partial t} (i_l W) + \frac{\rho_b}{\rho_g} \frac{\partial i_s}{\partial t} + v_b \frac{\partial}{\partial x} (i_{v,i} Y + i_g) = 0$$
(7)

The energy balance for the particles is more simple, because there is only vapor diffusion and it is given by:

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_{x_0}^{x_1} \left((1 - \epsilon_b) \epsilon_p \left(i_g \, \rho_g + i_{v,\pi} \, \rho_{v,\pi} \right) + \, i_l \, \rho_l + i_s \, \rho_b \right) \mathcal{A}_x \, \mathrm{d}x = \\ = \int_{x_0}^{x_1} \left(i_{v,f} \, h_m \, \rho_g \left(Y - Y_p \right) + \, h_h \left(T - T_p \right) \right) \, \mathcal{S} \, \mathcal{A}_x \, \mathrm{d}x \quad (8)$$

Lastly, simplification yields:

$$(1 - \epsilon_b)\epsilon_p \frac{\partial}{\partial t}(i_g + i_{v,\pi}Y_p) + \frac{\rho_b}{\rho_g}\frac{\partial}{\partial t}(i_lW) + \frac{\rho_b}{\rho_g}\frac{\partial i_s}{\partial t} = \left(i_{v,f}h_m(Y - Y_p) + \frac{h_h}{\rho_g}(T - T_p)\right)\mathcal{S}$$
(9)

Subtracting equation (9) from (7) yields a energy balance for the process gas flow:

$$\epsilon_b \frac{\partial}{\partial t} (i_g + i_{v,i} Y) + v_b \frac{\partial}{\partial x} (i_g + i_{v,i} Y) = -\left(i_{v,f} h_m \left(Y - Y_p\right) + \frac{h_h}{\rho_g} \left(T - T_p\right)\right) \mathcal{S}$$
(10)

3.3 Simplified Energy Balances in Terms of Temperatures

Defining dry-basis specific heats, an effective (apparent) density and introducing the heats of sorption, respectively as:

$$\rho_g \tilde{c} = \rho_g \left(c_{p_g} + Y c_{p_v} \right) \tag{11}$$

$$\rho_e \,\tilde{c}_e = \rho_b \left(c_s + c_l \, W \right) + \rho_g \left(c_{p_g} + Y_p \, c_{p_v} \right) \left(1 - \epsilon_b \right) \epsilon_p \tag{12}$$

$$\rho_e = \rho_b + (1 + \epsilon_b) \epsilon_p \rho_g \tag{13}$$

$$i_{sor}^{p} = i_{v,\pi} - i_{l}, \qquad i_{sor}^{i} = i_{v,i} - i_{v,\pi}, \qquad i_{sor} = i_{sor}^{p} + i_{sor}^{i} = i_{v,i} - i_{l}$$
 (14)

Using the definitions of specific heats for perfect gases and incompressible substances, expanding equations (9) and (10), using and multiplying the mass conservation equations (4) and (5) by $i_{v,\pi}$ and $i_{v,i}$, for particles and process gas flow equations, respectively, yields:

$$\frac{\rho_e}{\rho_g} \tilde{c}_e \frac{\partial T_p}{\partial t} = \left(\varphi \, i^i_{sor} \, h_m \left(Y - Y_p\right) \, + \, \frac{h_h}{\rho_g} \left(T - T_p\right)\right) \, S \, + \, \frac{\rho_b}{\rho_g} \, i^p_{sor} \, \frac{\partial W}{\partial t} \tag{15}$$

$$\tilde{c}\left(\epsilon_b \frac{\partial T}{\partial t} + v_b \frac{\partial T}{\partial x}\right) = \left((1-\varphi)i^i_{sor}h_m\left(Y-Y_p\right) - \frac{h_h}{\rho_g}\left(T-T_p\right)\right)S$$
(16)

where φ is the fraction of i_{sor}^i that contributes directly to heating (or cooling) the process gas flow; conversely, $1 - \varphi$ is the fraction of i_{sor}^i that contributes directly to heating (or cooling) the adsorbent particles.

4. NORMALIZATION

4.1 Dimensionless groups

 t^*

The first dimensionless groups are the dimensionless dependent and independent variables involved in the studied problem:

$$T_{p}^{*} = \frac{(T_{p} - T_{ref})}{\Delta T}, \qquad T^{*} = \frac{(T - T_{ref})}{\Delta T}, \qquad Y_{p}^{*} = \frac{Y_{p}}{Y_{max}}, \qquad Y^{*} = \frac{Y}{Y_{max}}, \qquad W^{*} = \frac{W}{W_{max}}$$
(17)

$$=\frac{t}{\tau}, \qquad x^* = \frac{x}{L} \tag{18}$$

where $\Delta T = T_{\text{max}} - T_{\text{min}}$ and $T_{\text{ref}} = T_{\text{min}}$.

The next groups are the dimensionless parameters, such as the dimensionless dwell time (residence period), volumetric and heat capacities ratios, dimensionless numbers of transfer units, dimensionless concentration of water in the adsorbent, dimensionless heat of adsorption and heat capacity ratios, expressed respectively as:

$$\tau_{res} = \frac{L}{v_b} \epsilon_b, \qquad \tau_{res}^* = \frac{\tau_{res}}{\tau}, \qquad \mathbf{V}_r = \frac{\mathcal{V}}{\tau}, \qquad \mathbf{V} = v_b \mathcal{A}_x, \qquad \mathbf{C}_r = \frac{c_b \rho_b L A_x}{\tau}$$
(19)

$$C = \rho_g v_b A_x c_{p_g}, \qquad \mathcal{V}_r^* = \frac{\mathcal{V}_r}{\mathcal{V}}, \qquad \mathcal{C}_r^* = \frac{\mathcal{C}_r}{\mathcal{C}}, \qquad \mathcal{X}_i = \frac{\tilde{c}}{c_{p_g}}, \qquad \mathcal{X}_p = \frac{\rho_e \, \tilde{c_e}}{\rho_b \, c_b}, \qquad \mathcal{N}_{tu}^h = \frac{h_h \, \mathcal{A}_s}{\mathcal{C}} \tag{20}$$

$$N_{tu}^{m} = \frac{h_{m} \mathcal{A}_{s}}{V}, \qquad \Omega = \frac{\rho_{b}}{\rho_{g}} \frac{W_{\max}}{Y_{\max}}, \qquad i_{sor}^{p*} = i_{sor}^{p} \frac{\rho_{g} Y_{\max}}{\rho_{b} c_{b} \Delta T}, \qquad i_{sor}^{i*} = i_{sor}^{i} \frac{\rho_{g} Y_{\max}}{\rho_{b} c_{b} \Delta T}$$
(21)

4.2 Dimensionless governing equations

For the mass balance of the particles, the dimensionless equation yields:

$$\Omega \operatorname{V}_{r}^{*} \frac{\partial W^{*}}{\partial t^{*}} + (1 - \epsilon_{b}) \epsilon_{p} \operatorname{V}_{r}^{*} \frac{\partial Y_{p}^{*}}{\partial t^{*}} = \operatorname{N}_{tu}^{m} (Y^{*} - Y_{p}^{*})$$

$$\tag{22}$$

For the mass balance of the process gas flow, the dimensionless equation is written as:

$$\tau_{\rm res}^* \frac{\partial Y^*}{\partial t^*} + \frac{\partial Y^*}{\partial x^*} = N_{tu}^m \left(Y_p^* - Y^* \right) \tag{23}$$

For the energy balance for the particles, the dimensionless equation is given by:

$$\chi_p \frac{\partial T_p^*}{\partial t^*} = \varphi \frac{\mathcal{N}_{tu}^m}{\mathcal{V}_r^*} i_{sor}^{i*} \left(Y^* - Y_p^*\right) + \frac{\mathcal{N}_{tu}^h}{\mathcal{C}_r^*} \left(T^* - T_p^*\right) + \Omega \, i_{sor}^{p*} \frac{\partial W^*}{\partial t^*} \tag{24}$$

For the energy balance of the process gas flow, the dimensionless equation results:

$$\chi_i \left(\tau_{\text{res}}^* \frac{\partial T^*}{\partial t^*} + \frac{\partial T^*}{\partial x^*} \right) = (\varphi - 1) \operatorname{N}_{tu}^m \frac{\operatorname{C}_r^*}{\operatorname{V}_r^*} i_{sor}^{i*} \left(Y_p^* - Y^* \right) + \operatorname{N}_{tu}^h \left(T_p^* - T^* \right)$$
(25)

4.3 Performance Assessment

The performance of the dehydration process is assessed by the dehydration effectiveness (η_m) and it has been employed for assessing the performance of desiccant wheels (Nóbrega and Brum, 2011; Niu and Zhang, 2002). The formulation comprises a measure of the level of dehydration of the gas stream with the ideal parameter $Y_{\text{out,min}}^* = 0$:

$$\eta_m = \frac{(Y_{\rm in}^* - Y_{\rm out}^*)}{(Y_{\rm in}^* - Y_{\rm out,min}^*)}$$
(26)

5. RESULTS AND DISCUSSION

To solve the equations, it was used the Finite Volume Method (Patankar, 1980) with a uniformly spaced mesh following the methodology presented in Sphaier and Worek (2009), and the entire implementation was done in the *Mathematica* system (Wolfram, 2003). To express the relationship between the quantity adsorbed W, humidity and temperature of gas in equilibrium with this phase, we used the following adsorption isotherm:

$$W^*\left(T_p^*, Y_p^*\right) = \frac{1}{1 - r + r/\phi}, \qquad \phi = \phi\left(T_p, Y_p\right) = \frac{Y_p}{r_M^{v,g} + Y_p} \frac{p_{\rm op}}{p_{\rm vs}\left(T_p\right)}, \qquad r_M^{v,g} = \frac{M_v}{M_g} \tag{27}$$

where ϕ is the relative humidity of natural gas presented in references (ASHRAE, 2005), r is the separation factor of the adsorption isotherm (Basmadjian, 1997) and $r_M^{v,g}$ is the relation between vapor and dry natural gas molecular masses (Friend *et al.*, 1989; Kohl and Nielsen, 1997).

The density of the gas was calculated assuming pure methane gas with ideal gas behavior. Table 1 presents the numerical data (Çengel and Boles, 1998) used in the simulations assuming silica gel as solid dehydration medium (Hubard, 1954).

Parameters	Symbols	Values
Minimum Temperature (K)	T_{\min}	300
Specific Mass of Methane (kg/m ³)	$ ho_g$	$5.512 \cdot 10^{-6} p_{op}$
Specific Heat of Methane (J/kg·K)	c_{p_a}	2253.7
Specific Heat of Adsorbent (J/kg·K)	c_b, c_s	921
Specific Heat of Liquid Water (J/kg·K)	c_l	4180
Specific Heat of Water Vapor (J/kg·K)	c_{p_n}	1872.3
Maximum Concentration of Water on Adsorbent (kg/kg)	$W_{\rm max}$	0.45
Molecular Mass of Methane (kg/kmol)	M_{g}	16.043
Molecular Mass of Water (kg/kmol)	M_v	18.015
Methane Gas Constant (J/kg·K)	R_{g}	518.35
Particle Porosity	ϵ_p	0.366
Bed Porosity	ϵ_b	0.3
Maximum Concentration of Vapor (g/kg)	$Y_{\rm max}$	32.1
Adsorption Heat (J/kg H ₂ O)	i_{sor}	$2.7 imes 10^6$
Heat Capacity Ratio	C_r^*	10^{-1}
Volumetric Capacity Ratio	\mathbf{V}_r^*	$570^{-1} \ \mathrm{C}_r^*$
Dimensionless Period of Residence	$ au_{ m res}^*$	$0.3 V_r^*$
Dimensionless Concentration of Water in Adsorbent	Ω	19557
Dimensionless Heat of Adsorption	i_{sor}^{i*}	0

Table 1. Values Used in the Numerical Solution

The dimensionless inlet values, Y_{in}^* and T_{in}^* , can assume different values for each process, but for the studied cases it was assumed $Y_{in}^* = 1$ and $T_{in}^* = 0$ at adsorption process and $Y_{in}^* = 1$ and $T_{in}^* = 1$ at regeneration process, as observed in figure 1. For the adsorption processes, the inlet condition describes that the fluid is totally saturated with water and the temperature is the minimum established. For the regeneration process, the absolute humidity of the gas is the same as the adsorption process inlet, however the temperature is the maximum stipulated.

For the first studied case with separation factor r = 0.1 and maximum temperature $T_{\text{max}} = 400$ K as shown in figure 2, the dehydration effectiveness (η_m) increases with pressure (p) and number of transfer unit (N_{tu}^h) . Nearly by $N_{tu}^h = 4$, it is observed a non increasing behavior of η_m , so one can considered it as a best point operation. At some pressure value there is not significant η_m increase, as result the pressure can be limited to an operation value with minimum waste of compression power.

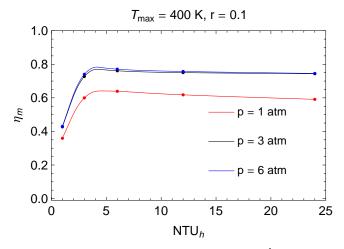


Figure 2. Variation of η_m with respect to N_{tu}^h for r = 0.1

In figure 3 with separation factor r = 1 and $T_{\text{max}} = 400$ K, the η_m also increases with p and N_{tu}^h . About $N_{tu}^h = 4$,

it is observed a non increasing behavior of η_m , so one can considered it as a good region of operation. At some pressure value there is not significant η_m increase, as result the pressure can be limited to an operation value with minimum waste of compression power. Relatively to the case shown in figure 3, it is identified a better η_m response to higher pressures, thus a solid desiccant with r = 1 is foremost to operate.

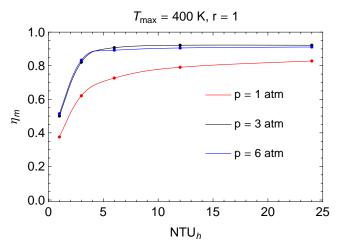


Figure 3. Variation of η_m with respect to N_{tu}^h for r = 1

For the last case as shown in figure 4 with separation factor r = 10 and $T_{\text{max}} = 400$ K, it is noted one more time that the η_m increases with p and N_{tu}^h . About $N_{tu}^h = 5$, it is observed a constant behavior of η_m , so one can considered it as a good operation parameter. In this case, the pressure variation has more significance than the last two cases, however for lower pressures the η_m is not relatively fine.

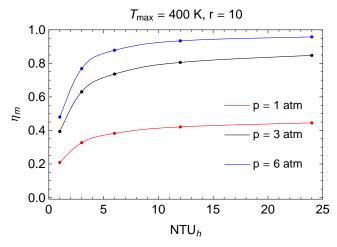


Figure 4. Variation of η_m with respect to N_{tu}^h for r = 10

6. CONCLUSIONS

This study presented a simple mathematical model for simulating the process of natural gas dehydration using solid desiccants. A system of one-dimensional governing equations for heat and mass transfer within the adsorbent material were obtained. Dimensionless groups associated to this problem were presented and the formulation was normalized using these groups. A numerical solution of the problem was implemented using the Finite Volumes Method combined with the Method of Lines. Then, simulation results were carried-out to illustrate the effect of varying the operation pressure on the dehydration effectiveness. The results show that for the simulated conditions, bigger pressures results in better

dehydration effectiveness. For most studied cases, there are betters regions to operate such as between $N_{tu}^{h} = 4$ and $N_{tu}^{h} = 5$. For higher operation pressures, it is better to use solid desiccants with higher r to obtain greater η_m , however for lower pressures, it is recommended to use lower r to gain greater η_m . In spite of the relevance of this study, the results are still preliminary and future research should definitely be conducted.

7. ACKNOWLEDGEMENTS

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