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# HETP Evaluation of Structured and Randomic Packing Distillation Column 

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## 1. Introduction

Packed columns are equipment commonly found in absorption, distillation, stripping, heat exchangers and other operations, like removal of dust, mist and odors and for other purposes. Mass transfer between phases is promoted by their intimate contact through all the extent of the packed bed. The main factors involving the design of packed columns are mechanics and equipment efficiency. Among the mechanical factors one could mention liquid distributors, supports, pressure drop and capacity of the column. The factors related to column efficiency are liquid distribution and redistribution, in order to obtain the maximum area possible for liquid and vapor contact (Caldas and Lacerda, 1988).
These columns are useful devices in the mass transfer and are available in various construction materials such as metal, plastic, porcelain, ceramic and so on. They also have good efficiency and capacity, moreover, are usually cheaper than other devices of mass transfer (Eckert, 1975).
The main desirable requirements for the packing of distillation columns are: to promote a uniform distribution of gas and liquid, have large surface area (for greater contact between the liquid and vapor phase) and have an open structure, providing a low resistance to the gas flow. Packed columns are manufactured so they are able to gather, leaving small gaps without covering each other. Many types and shapes of packing can satisfactorily meet these requirements (Henley and Seader, 1981).
The packing are divided in random - randomly distributed in the interior of the column and structured - distributed in a regular geometry. There are some rules which should be followed when designing a packed column (Caldas and Lacerda, 1988):
a. The column should operate in the loading region ( 40 to $80 \%$ flooding), which will assure the best surface area for the maximum mass transfer efficiency;
b. The packing size (random) should not be greater than $1 / 8$ the column diameter;
c. The packing bed is limited to 6 D (Raschig rings or sells) or 12D for Pall rings. It is not recommended bed sections grater than 10 m ;
d. Liquid initial distribution and its redistribution at the top of each section are very important to correct liquid migration to the column walls.
A preliminary design of a packed column involves the following steps:

1. Choice of packing;
2. Column diameter estimation;
3. Mass transfer coefficients determination;
4. Pressure drop estimation;
5. Internals design.

This chapter deals with column packing efficiency, considering the main studies including random and structured packing columns. In packed columns, mass transfer efficiency is related to intimate contact and rate transfer between liquid and vapor phases. The most used concept to evaluate the height of a packed column, which is related to separation efficiency, is the HETP (Height Equivalent to Theoretical Plate), defined by the following equation:

$$
\begin{equation*}
Z=(H E T P) \cdot(N) \tag{1}
\end{equation*}
$$

in which $Z$ is the height of the packed bed necessary to obtain a separation equivalent to $N$ theoretical stages (Caldas and Lacerda, 1988).
Unfortunately, there are only a few generalized methods available in the open literature for estimating the HETP. These methods are empirical and supported by the vendor advice. The performance data published by universities are often obtained using small columns and with packing not industrially important. When commercial-scale data are published, they usually are not supported by analysis or generalization (Vital et al., 1984). Several correlations and empirical rules have been developed for HETP estimation in the last 50 years. Among the empirical methods, there is a rule of thumb for traditional random packing that says
HETP = column diameter

That rule can be used only in small diameter columns (Caldas and Lacerda, 1988).
The empirical correlation of Murch (1953) cited by Caldas and Lacerda (1988) is based on HETP values published for towers smaller than 0.3 m of diameter and, in most cases, smaller than 0.2 m . The author had additional data for towers of $0.36,0.46$ and 0.76 m of diameter. The final correlation is

$$
\begin{equation*}
H E T P=K_{1} G^{K_{2}} D^{K_{3}} Z^{1 / 3}\left(\frac{\alpha \mu_{L}}{\rho_{L}}\right) \tag{3}
\end{equation*}
$$

$K_{1}, K_{2}$ and $K_{3}$ are constants that depend on the size and type of the packing.
Lockett (1998) has proposed a correlation to estimate HETP in columns containing structured packing elements. It was inspired on Bravo et al.'s correlation (1985) in order to develop an empirical relation between HETP and the packing surface area, operating at $80 \%$ flooding condition (Caldas and Lacerda, 1988):

$$
\begin{equation*}
\text { HETP }=\frac{\left(4.82\left(\rho_{L}-\rho_{G}\right)^{0.5} \mu_{r}^{-0.06}\right)}{\alpha} \tag{4}
\end{equation*}
$$

in which

$$
\begin{equation*}
\alpha=a_{p}\left[\left(1+0.78 e^{0.00058 a_{p}}\right)\left(\frac{\rho_{G}}{\rho_{L}}\right)^{0.25}\right]^{2} \tag{5}
\end{equation*}
$$

According to the double film theory, HETP can be evaluated more accurately by the following expression (Wang et al., 2005):

$$
\begin{equation*}
H E T P=\frac{\ln \lambda}{\lambda-1}\left[\frac{u_{G s}}{k_{G} a_{e}}+\lambda \frac{u_{L s}}{k_{L} a_{e}}\right] \tag{6}
\end{equation*}
$$

Therefore, the precision to evaluate HETP by equation (6) depends on the accuracy of correlations used to predict the effective interfacial area and the vapor and liquid mass transfer coefficients. So, we shall continue this discussion presenting the most used correlations for wetted area estimation, both for random and structured packed columns. Wang et al. (2005) also presented a complete discussion about the different correlations mostly used for random and structured packing.

## 2. Literature review

The literature review will be divided in two sections, treating and analyzing separately random and structured distillation columns as the correlations for the effective area and HETP evaluation.

### 2.1 Part A: performance of random packing

Before 1915, packed columns were filled with coal or randomly with ceramic or glass shards. This year, Fredrick Raschig introduced a degree of standardization in the industry. Raschig rings, together with the Berl saddles, were the packing commonly used until 1965. In the following decade, Pall rings and some more exotic form of saddles has gained greater importance (Henley and Seader, 1981). Pall rings are essentially Raschig rings, in which openings and grooves were made on the surface of the ring to increase the free area and improve the distribution of the liquid. Berl saddles were developed to overcome the Raschig rings in the distribution of the liquid. Intalox saddles can be considered as an improvement of Berl saddles, and facilitated its manufacture by its shape. The packing Hypac and Super Intalox can be considered an improvement of Pall rings and saddles Intalox, respectively (Sinnott, 1999). In Figure 1, the packing are illustrated and commented.
The packing can be grouped into generations that are related to the technological advances. The improvements cited are from the second generation of packing. Today, there are packing of the fourth generation, as the Raschig super ring (Darakchiev \& Semkov, 2008).
Tests with the objective to compare packing are not universally significant. This is because the efficiency of the packing does not depend, exclusively, on their shape and material, but other variables, like the system to be distilled. This means, for example, that a packing can not be effective for viscous systems, but has a high efficiency for non-viscous systems. Moreover, the ratio of liquid-vapor flow and other hydrodynamic variables also must be considered in comparisons between packing. The technical data, evaluated on packing, are, generally, the physical properties (surface area, free area, tensile strength, temperature and chemical stability), the hydrodynamic characteristics (pressure drop and flow rate allowable) and process efficiency (Henley and Seader, 1981). This means that Raschig rings can be as efficient as Pall rings, depending on the upward velocity of the gas inside the column, for example. These and other features involving the packing are extensively detailed in the study of Eckert (1970).


Ref: Henley \& Seader (1981)
Fig. 1. Random packing: (a) plastic pall rings. (b) metal pall rings (Metal Hypac). (c) Raschig rings. (d) Intalox saddles. (e) Intalox saddles of plastic. (f) Intalox saddles
In literature, some studies on distillation show a comparison between various types of random and structured packing. Although these studies might reveal some tendency of the packing efficiency for different types and materials, it is important to emphasize that they should not generalize the comparisons.
Cornell et al. (1960) published the first general model for mass transfer in packed columns. Different correlations of published data of $H_{L}$ and $H_{V}$, together with new data on industrial scale distillation columns, were presented to traditional packing, such as Raschig rings and Berl saddles, made of ceramic. Data obtained from the experimental study of $H_{L}$ and $H_{V}$ were analyzed and correlated in order to project packed columns. The heights of mass transfer for vapor and liquid phases, are given by:

$$
\begin{gather*}
H_{V}=\frac{\psi \cdot S_{C V}^{0,5}}{\left(G_{L} \cdot f_{1} \cdot f_{2} \cdot f_{3}\right)^{n}} \cdot\left(\frac{d_{C}}{12}\right)^{m} \cdot\left(\frac{Z}{10}\right)^{\frac{1}{3}}  \tag{7}\\
H_{L}=\phi \cdot C_{f L} \cdot\left(\frac{Z}{10}\right)^{0,15} \cdot S_{C L}^{0,5} \tag{8}
\end{gather*}
$$

which:

$$
\begin{align*}
& f_{1}=\left(\frac{\mu_{L}}{\mu_{W}}\right)^{0,16}  \tag{9}\\
& f_{2}=\left(\frac{\rho_{W}}{\rho_{L}}\right)^{1,25}  \tag{10}\\
& S_{C V}=\frac{\mu_{G}}{\rho_{G} \cdot D_{G}}  \tag{11}\\
& S_{C L}=\frac{\mu_{L}}{\rho_{L} \cdot D_{L}} \tag{12}
\end{align*}
$$

In the $f$ factors, the liquid properties are done in the same conditions of the column and the water properties are used at $20^{\circ} \mathrm{C}$. The parameters $n$ and $m$ referred to the packing type, being 0.6 and 1.24 , respectively, for the Raschig rings. $C_{f L}$ represents the approximation coefficient of the flooding point for the liquid phase mass transfer. The values of $\varphi$ and $\Psi$ are packing parameters for the liquid and vapor phase mass transfer, respectively, and are graphically obtained. In this correlation, some variables don't obey a single unit system and therefore need to be specified: $d c(\mathrm{in}), \mathrm{Z}(\mathrm{ft}), H(\mathrm{ft}), G\left(\mathrm{lbm} / \mathrm{h} . \mathrm{ft}^{2}\right)$.
Onda et al. (1968 a, b) presented a new model to predict the global mass transfer unit. In this method, the transfer units are expressed by the liquid and vapor mass transfer coefficients:

$$
\begin{gather*}
H_{V}=\frac{G_{V}}{k_{V} \cdot a_{W} \cdot P \cdot M_{V}}  \tag{13}\\
H_{L}=\frac{G_{L}}{k_{L} \cdot a_{W} \cdot \rho_{L}} \tag{14}
\end{gather*}
$$

In which:

$$
\begin{gather*}
k_{V} \cdot\left(\frac{R \cdot T}{a_{P} \cdot D_{V}}\right)=\Gamma \cdot\left(\frac{G_{V}}{a_{P} \cdot \mu_{V}}\right)^{0,7} \cdot\left(S_{C V}\right)^{\frac{1}{3}} \cdot\left(a_{P} \cdot d_{P}\right)^{-2}  \tag{15}\\
k_{L} \cdot\left(\frac{\rho_{L}}{g \cdot \mu_{L}}\right)^{\frac{1}{3}}=0,0051 \cdot\left(\frac{G_{L}}{a_{w} \cdot \mu_{L}}\right)^{\frac{2}{3}} \cdot\left(S_{C L}\right)^{-\frac{1}{2}} \cdot\left(a_{P} \cdot d_{P}\right)^{0,4} \tag{16}
\end{gather*}
$$

where $\Gamma$ is a constant whose values can vary from 5.23 (normally used) or 2 , if the packing are Raschig rings or Berl saddles with dimension or nominal size inferior to 15 mm .
It can be noted, in these equations, the dependence of the mass transfer units with the wet superficial area. It is considered, in this model, that the wet area is equal to the liquid-gas interfacial area that can be written as

$$
\begin{equation*}
a_{W}=a_{p} \cdot\left\{1-\exp \left[-1,45 \cdot \operatorname{Re}_{L}^{0,1} \cdot \mathrm{Fr}_{L}^{-0,05} \cdot W e_{L}^{0,2} \cdot\left(\frac{\sigma}{\sigma_{\mathrm{C}}}\right)^{-0,75}\right]\right\} \tag{17}
\end{equation*}
$$

where:

$$
\begin{gather*}
\operatorname{Re}_{L}=\frac{G_{L}}{a_{P} \cdot \mu_{L}}  \tag{18}\\
W e_{L}=\frac{G_{L}^{2}}{a_{P} \cdot \sigma \cdot \rho_{L}}  \tag{19}\\
F r_{L}=\frac{a_{P} \cdot G_{L}^{2}}{g \cdot \rho_{L}^{2}} \tag{20}
\end{gather*}
$$

The ranges in which the equation should be used are: $0.04<R e_{L}<500 ; 1.2 \times 10^{-8}<W e_{L}<0.27$; $2.5 \times 10^{-9}<F r_{L}<1.8 \times 10^{-2} ; 0.3<\left(\sigma_{d} / \sigma\right)<2$.
The equation for the superficial area mentioned can be applied, with deviations of, approximately, $20 \%$ for columns packed with Raschig rings, Berl saddles, spheres, made of ceramic, glass, certain polymers and coatted with paraffin.
Bolles and Fair (1982) compiled and analyzed a large amount of performance data in the literature of packed beds, and developed a model of mass transfer in packed column. Indeed, the authors expanded the database of Cornell et al. (1960) and adapted the model to new experimental results, measured at larger scales of operation in another type of packing (Pall rings) and other material (metal). The database covers distillation results in a wide range of operating conditions, such as pressures from 0.97 to 315 psia and column diameters between 0.82 to 4.0 ft . With the inclusion of new data, adjustments were needed in the original model and the values of $\varphi$ and $\Psi$ had to be recalculated. However, the equation of Bolles and Fair model (1982) is written in the same way that the model of Cornell et al. (1960). The only difference occurs in the equation for the height of mass transfer to the vapor phase, just by changing the units of some variables:

$$
\begin{equation*}
H_{V}=\frac{\psi \cdot S_{C V}^{0,5}}{\left(3600 \cdot G_{L} \cdot f_{1} \cdot f_{2} \cdot f_{3}\right)^{n}} \cdot d_{C}^{\prime} \cdot\left(\frac{Z}{10}\right)^{\frac{1}{3}} \tag{21}
\end{equation*}
$$

In this equation, $d^{\prime}{ }_{C}$ is the adjusted column diameter, which is the same diameter or 2 ft , if the column presents a diameter higher than that.
Unlike the graphs for estimating the values of $\varphi$ and $\Psi$, provided by Cornell et al. (1960), where only one type of material is analyzed (ceramic) and the percentage of flooding, required to read the parameters, is said to be less than $50 \%$ in the work of Bolles and Fair (1982), these graphics are more comprehensive, firstly because they include graphics for Raschig rings, Berl saddles and metal Pall rings, and second because they allow variable readings for different flooding values.
The flooding factor, necessary to calculate the height of a mass transfer unit in the Bolles and Fair (1982) model, is nothing more than the relation between the vapor velocity, based on
the superficial area of the column, and the vapor velocity, based on the superficial area of the column at the flooding point. The Eckert model (1970) is used for the determination of these values. The authors compared the modified correlation with the original model and with the correlation of Onda et al. (1968 a, b), concluding that the lower deviations were obtained by the proposed model, followed by the Cornell et al. (1960) model and by the Onda et al. (1968 a, b) model.
Bravo and Fair (1982) had as objective the development of a general project model to be applied in packed distillation columns, using a correlation that don't need validation for the different types and sizes of packing. Moreover, the authors didn't want the dependence on the flooding point, as the model of Bolles and Fair (1982). For this purpose, the authors used the Onda et al. (1968 a, b) model, with the database of Bolles and Fair (1982) to give a better correlation, based on the effective interfacial area to calculate the mass transfer rate. The authors suggested the following equation:

$$
\begin{equation*}
a_{e}=\frac{\left(a_{e} \cdot H_{V}+\lambda \cdot a_{e} \cdot H_{L}\right)}{H_{O V}} \tag{22}
\end{equation*}
$$

Evidently, the selection of $k_{V}$ e $k_{L}$ models is crucial, being chosen by the authors the models of Shulman et al. (1955) and Onda et al. (1968a, b), since they correspond to features commonly accepted. The latter equation has been written in equations 23 and 24 . For the first, we have:

$$
\begin{align*}
\frac{k_{V} \cdot \rho_{V} \cdot R T}{G_{V}} & =1.195 \cdot\left[\frac{d_{p}^{\prime} \cdot G_{V}}{\mu_{V} \cdot(1-\varepsilon)}\right]^{-0.36} \cdot\left(S_{C V}\right)^{-\frac{2}{3}}  \tag{23}\\
\frac{k_{L} \cdot d_{p}^{\prime}}{D_{L}} & =25.1 \cdot\left[\frac{d_{p}^{\prime} \cdot G_{L}}{\mu_{L}}\right]^{0.45} \cdot\left(S_{C L}\right)^{0.50} \tag{24}
\end{align*}
$$

The database used provided the necessary variables for the effective area calculation by the both methods. These areas were compared with the known values of the specific areas of the packing used. Because of that, the Onda et al. (1968 a, b) model was chosen to provide moderate areas values, beyond cover a large range of type and size packing and tested systems.
The authors defined the main points that should be taken in consideration by the new model and tested various dimensional groups, including column, packing and systems characteristics and the hydrodynamic of the process. The better correlation, for all the systems and packing tested is given by:

$$
\begin{equation*}
\frac{a_{e}}{a_{p}}=0.498 \cdot\left(\frac{\sigma^{0.5}}{Z^{0.4}}\right) \cdot\left(C a_{L} \cdot \operatorname{Re}_{V}\right)^{0.392} \tag{25}
\end{equation*}
$$

which:

$$
\begin{equation*}
C a_{L}=\frac{\mu_{L} \cdot G_{L}}{\rho_{L} \cdot \sigma \cdot g_{C}} \tag{26}
\end{equation*}
$$

$$
\begin{equation*}
\operatorname{Re}_{V}=\frac{6 \cdot G_{V}}{a_{P} \cdot \mu_{V}} \tag{27}
\end{equation*}
$$

Recently, with the emergence of more modern packing, other correlations to predict the rate of mass transfer in packed columns have been studied. Wagner et al. (1997), for example, developed a semi-empirical model, taking into account the effects of pressure drop and holdup in the column for the Nutter rings and IMTP, CMR and Flaximax packing. These packing have higher efficiency and therefore have become more popular for new projects of packed columns today. However, for the traditional packing, according to the author, only correlations of Cornell et al. (1960), Onda et al. (1968a, b), Bolles and Fair (1982) and Bravo and Fair (1982), presented have been large and viable enough to receive credit on commercial projects for both applications to distillation and absorption.
Berg et al. (1984) questioned whether the extractive distillation could be performed in a packed distillation column, or only columns with trays could play such a process. Four different packing were used and ten separation agents were applied in the separation of ethyl acetate from a mixture of water and ethanol, which results in a mixture that has three binary azeotropes and a ternary. A serie of runs was made in a column of six glass plates, with a diameter of 3.8 cm , and in two packed columns. Columns with Berl saddles and Intalox saddles (both porcelain and 1.27 cm ) had 61 cm long and 2.9 cm in diameter. The columns with propellers made of Pyrex glass and with a size of 0.7 cm , and Raschig rings made of flint glass and size 0.6 cm , were 22.9 cm long and 1.9 cm in diameter. The real trays in each column were determined with a mixture of ethyl benzene and $m$-xylene. The cell, fed with the mixture, remained under total reflux at the bubble point for an hour. After, the feed pump was switched on and the separating agent was fed at $90^{\circ} \mathrm{C}$ at the top of the column. Samples from the top and bottom were analyzed every half hour, even remain constant, two hours or less. The results showed, on average, than the packed column was not efficient as the columns of plates for this system. The best packing for this study were, in ascending order, glass helices, Berl saddles, Intalox saddles and Raschig rings. The columns with sieve plates showed the best results. Propellers glass and Berl saddles were not as effective as the number of perforated plates and Intalox saddles and Raschig rings were the worst packing tested. When the separating agent was 1,5 -pentanediol, the tray column showed a relative volatility of ethyl acetate/ethanol of 3.19. While the packed column showed 2.32 to Propeller glass, 2.08 for Berl saddles, 2.02 for Intalox saddles and 2.08 for Raschig rings.
Through the years, several empirical rules have been proposed to estimate the packing efficiency. Most of the correlations and rules are developed for handles and saddle packing. Vital et al. (1984) cited several authors who proposed to develop empirical correlations for predicting the efficiency of packed columns (Furnas \& Taylor, 1940; Robinson \& Gilliand, 1950; Hands \& Whitt, 1951; Murch, 1953; Ellis, 1953 and Garner, 1956).
According to Wagner et al. (1997), the HETP is widely used to characterize the ability of mass transfer in packed column. However, it is theoretically grounded in what concerns the mass transport between phases. Conversely, the height of a global mass transfer, HOV, is more appropriate, considering the mass transfer coefficient ( $k$ ) of the liquid phase (represented by subscript L) and vapor (represented by subscript V) individually. Thus, the knowledge of the theory allows the representation:

$$
\begin{equation*}
H_{O V}=H_{V}+\lambda \cdot H_{L} \tag{28}
\end{equation*}
$$

$$
\begin{gather*}
H_{V}=\frac{G_{V}}{k_{V} \cdot a_{e} \cdot P \cdot M_{V}}  \tag{29}\\
H_{L}=\frac{G_{L}}{k_{L} \cdot a_{e} \cdot \rho_{L}} \tag{30}
\end{gather*}
$$

The effective interfacial mass transfer area, in a given system, is considered equal to the liquid and vapor phases, as is the area through which mass transfer occurs at the interface. It is important to also note that $a_{e}$ is not composed only by the wet surface area of the packing $\left(a_{W}\right)$, but throughout the area that allows contact between the liquid and vapor phases (Bravo and Fair, 1982). This area can be smaller than the global interfacial area, due to the existence of stagnant places, where the liquid reaches saturation and no longer participate in the mass transfer process. Due to this complicated physical configuration, the effective interfacial area is difficult to measure directly. The authors proposed a new model using high-efficiency random packing as IMTP, CMR, Fleximac and Nutter. The final model became

$$
\begin{equation*}
\text { HETP }=C_{p k} \frac{Z^{0.5}}{a_{p}}\left(\frac{\pi(\epsilon-h) u_{V}}{4 D_{V}}\right)^{0.5}\left[1+\left(\frac{h D_{V} M_{L} \rho_{V}}{(c-h) D_{L} M_{V} \rho_{L}} \frac{V}{L}\right)^{0.5}\right]\left[\left(\frac{1-\epsilon+h}{1-c}\right)^{2 / 3}-1\right]^{-1} \tag{31}
\end{equation*}
$$

After the test using 326 experimental data, the predicted values of HETP showed a deviation less than $25 \%$ from the experimental results. It was observed that physical properties have a little effect on mass transfer.
Four binary systems were tested (cyclohexane-heptane, methanol-ethanol, ethylbenzenestyrene and ethanol-water) from different database and different packing types and sizes. The only packing parameter needed was a packing characteristic which has a value of 0.030 for a 2 in Pall and Raschig rings and about 0.050 for 2 in nominal size of the high efficiency packing investigated.
The theoretical relations between the mass transfer coefficient and a packing efficiency definition, are not easily obtained, in a general manner. This is due to the divergence between the mechanisms of mass transfer in a packed section and the concept of an ideal stage. The theoretical relation deduced, applied in the most simple and commonly situation is described as:

$$
\begin{equation*}
H E T P=H_{O V} \cdot \frac{\ln \lambda}{\lambda-1} \tag{32}
\end{equation*}
$$

Although validated only for the cases of dilute solutions, constant inclination of the equilibrium line, constant molar flow rates, binary systems and equimolar countercurrent diffusion, this equation has been applied to systems with very different conditions from these, and even for multicomponent systems (Caldas \& Lacerda, 1988).
The design of packed columns by the method of the height of a global mass transfer unit is an established practice and advisable. For this, it is necessary to know the height of the mass transfer unit for both liquid and for vapor phases.
$H_{L}$ values are usually experimentally obtained by absorption and desorption of a gas, slightly soluble, from a liquid film flowing over a packed tower, in a countercurrent mode with an air stream. Under these conditions, changes in gas concentration are neglected and
no resistance in the gas film is considered. The variables that affect the height of the liquid transfer unit are the height of the packed section, gas velocity, column diameter, the physical properties of liquid and the type and size of the packing.
The values of the height of a transfer unit of a gas film, $H_{V}$, need to be measured under the same conditions as the resistance of the liquid film is known. This can be done by the absorption of a highly soluble gas. An alternative method to determine $H_{V}$ involves the vaporization of a liquid, at constant temperature, within a gas stream. In this case, the resistance of the liquid film is zero and $H_{V}$ is equal to $H_{O V}$. The variables that affect the height of transfer unit of a gas film are the gas and liquid velocities, the physical properties of the gas, column diameter, the height, type and size of the packing (Cornell et al., 1960).
Linek et al. (2001) studied the hydraulic and mass transfer data measuring pressure drop, liquid hold-up, gas and liquid side volumetric mass transfer coefficients and the interfacial area for Rauschert-Metall-Sattel-Rings (RMSR) with 25,40 and 50 mm . The shape and characteristics of the studied packing corresponded with the metal Pall rings and Intalox packing of Norton. The distillation experiments were performed using the systems methanol-ethanol, ethanol-water and isooctane-toluene at atmospheric pressure in a column of diameter 0.1 m and a height of packing 1.67 m , operated under total reflux. The measured values of HETP were compared with those calculated for the different sizes of RMSR packing for the distillation systems. The calculated values differ by less than $\pm 15 \%$ from the experimental values, with the exception for the data obtained at extremely low gas flow rates in the system ethanol-methanol for which the respective difference reached $46 \%$. Figure 2, from the paper of Linek et al. (2001), shows the comparison of measured values of HETP with those calculated from absorption mass transfer data using the model described in Linek et al. (1995) cited by Linek et al. (2001).


Fig. 2. Comparison of measured values of HETP with those calculated from absorption mass transfer data (Linek et al., 2001)

Senol (2001) studied the performance of a randomly packed distillation column depending on the effective vapor-liquid interfacial area and the flood ratio. The analysis were mainly focused optimizing HETP and effective interfacial areas as function of the flood ratio estimated by Eckert flooding model (Eckert, 1970 cited by Senol, 2001). The experiments were done in a pilot scale column of 9 cm inside diameter randomly filled to a depth of 1.90 m with Raschig-type ceramic rings under atmospheric pressure. The runs were conducted to determine the capacity and efficiency at total reflux for several pressure drops. The efficiency tests were made using three packings of $6.25,9$ and 10.8 mm nominal sizes and binary systems like trichloroethylene/heptane, methylcyclohexane/toluene, heptane/toluene and benzene/toluene. The HETP was obtained by the Fenske equation. The efficiency results gave evidence of two critical factors, the flood ratio and the packing geometry that affects significantly the magnitude of effective interfacial area.
A working database of 2350 measurements (under total molar reflux), in the work of Piché et al. (2003), were extracted from the open literature to generate height equivalent to a theoretical plate (HETP) calculations, essential for the design of randomly packed distillation columns. According to the authors, the HETP approaches more a rule of thumb concept than an exact science and can be calculated as:

$$
\begin{equation*}
H E T P=\frac{\ln \left(m G^{\prime} / L^{\prime}\right)}{m G^{\prime} / L^{\prime}-1}\left(\frac{U_{G}}{k_{G} a_{w}}+m \frac{U_{L}}{k_{L} a_{w}}\right) \tag{33}
\end{equation*}
$$

The database used included 325 measurements on the interfacial area, 1100 measurements on the liquid-film coefficient $\left(k_{L} a_{w}\right), 361$ measurements on the gas-film coefficient $\left(k_{G} a_{w}\right)$, 1242 measurements on the liquid-overall coefficient ( $\mathrm{K}_{\mathrm{L}} \mathrm{a}_{\mathrm{w}}$ ) and 742 measurements on the gas-overall coefficient $\left(\mathrm{K}_{\mathrm{G}} \mathrm{a}_{\mathrm{w}}\right)$. The distillation database constituted 2357 HETP measurements taken from 22 different references, conducted at total molar reflux with standard binary mixtures (chlorobenzene-ethylbenzene, ethylbenzene-styrene, benzenetoluene, methanol/ethanol, trans-decalin/cis-decalin, ethanol-water, hexane-heptane, isopropanol-water, iso-octane-toluene, toluene-methylcyclohexane, cyclohexanecyclohexanol, o-xylene-p-xylene, benzene-1,1-dichloroethylene, trichloroethylene-n-heptane, n-heptane-toluene). All the systems were distilled using 24 varieties of packing. After the construction of a new model based on a neural network, the deviations were calculated and were better than the original model of Piche et al. (2002) cited in Piche et al. (2003). The minimum deviation of HETP was $21.3 \%$, including all the systems studied.
Darakchiev \& Semkov (2008) studied the rectification of ethanol with three types of modern random packings, IMTP, Raschig Super Rings and Ralu Flow, in conditions close to real conditions of industrial operation. The experiments were performed in high and medium concentrations. The experimental unit consisted of a column of internal diameter of 21.3 cm , made of stainless steel, with reboiler of $80000 \mathrm{~cm}^{3}$ of capacity and maximum resistance of 45 kW , condenser, pipes, devices for monitoring and measurement and a control panel. The column was built in separated sections and assembled by flanges. The packed section has a height of 2.8 m . To limit the damaging effect of preferential channels, reflectors rings were willing on 20 cm distance between the height of the packing. One type of disperser, a type of liquid distributor, with 21 holes of 3 mm with Teflon nozzle of 1.7 mm , was attached to the upper spine. To prevent clogging, a filter was placed before the distributor.
A diaphragm and a differential manometer were used to measure the discharge flow, which may be total or partial. The column was insulated by a layer of 50 mm glass fiber. The
experimental runs were done feeding $60000 \mathrm{~cm}^{3}$ of the solution to reboiler. The minimum liquid flow rate in the distributor needed to ensure good distribution of liquid in the column was obtained, experimentally, in $58000 \mathrm{~cm}^{3} / \mathrm{h}$, which required a minimum output of 13 kW . After equilibrium, samples were taken before and after the packing. A densimeter was used to determine the concentration of the samples, applying temperature corrections. Eight types of random packings were studied: four metal Raschig Super Rings, with dimensions of $1.27,1.52,1.78$ and 2.54 cm , a Raschig Super Ring, made of plastic, of 1.52 cm , two kinds of IMTP packing and a Ralu Flow of plastic. The results showed good efficiency of the packing in ethanol dehydration. The best packing tested was Raschig Super Ring in the smaller dimension, producing a HETP with 28 cm . Comparing metal to plastic, there was a $6 \%$ lower efficiency for plastic packing.
Larachi et al. (2008) proposed two correlations to evaluate the local gas or liquids side mass transfer coefficient and the effective gas-liquid interfacial area. The study was done using structured and random packing, testing 861 experiments for structured packings and 4291 experiments for random packings. In order to reconstruct HETP values from the mass transfer parameters, 1192 HETP experiments for random packings and 127 experiments for structured packings were evaluated. All the distillation experiments were done at total molar reflux with standard binary mixtures as chlorobenzene/ethylbenzene, ethylbenzene/styrene, benzene/toluene, methanol/ethanol, etc. All the physical chemical properties unavailable were predicted according to the rules of Reid et al. (1987). According to the neural network weights, the relative deviation was $29.2 \%$ for the random packing and $18.2 \%$ for the experiments done with structured packings.
Soares (2010) studied the ethanol concentration using different salts, $\mathrm{NaCl}, \mathrm{CaCl} 2, \mathrm{Ca}\left(\mathrm{NO}_{3}\right)_{2}$, sodium acetate and potassium acetate, in a packed column, with 5.9 cm of internal diameter and 37 cm of packed section height, with Raschig rings $(0.73 \mathrm{~cm})$ made of glass. The HETP was evaluated operating the distillation column at total reflux ratio. Despite of the less efficiency compared with the more modern packing, this type of packing presented the lowest costs. The correlations used in this work were Bravo \& Fair (1982), Bolles and Fair (1982) and Onda et al. (1968 a, b). The Cornell et al. (1960) correlation was not adopted, due to the fact that the model of Bolles and Fair (1982) is its improvement. All the physicalchemical properties were estimated by the different methods present in Reid et al. (1987). The thermodynamic modeling was done based on the work of Macedo et al. (1990), that introduced the Debye-Hückel term in UNIQUAC, to calculate the phase equilibrium for electrolytes. Two systems with different ethanol concentration were studied, 7 and $52^{\circ} \mathrm{GL}$. The better results of predicted HETP were obtained using the Onda et al. (1968 a, b) and with the Bolles and Fair (1982) correlations. The results predicted by the correlation of Bravo and Fair (1982) modified by Onda et al. (1968 a, b) were much higher than the experimental HETP. According to Caldas and Lacerda (1988), the maximum deviation is $27 \%$ using Raschig rings made of ceramic.
The choice for a distillation column is based on the cost and on the properties of the studied system. In the past, except for the columns with small diameters, the trays are adopted in the most of the distillation columns. However, the development of high efficient packing and the need for the improvement of the capacity, efficiency, and to reduce the pressure drop, has led to a more use of the packing columns in a large wide of applications in an industrial scale (Perry and Green, 1999).
The difference in cost and height, between the tray and packed columns, are not significant, if operating conditions are providing efficiency close to maximal. In general, trays are used
in large diameter columns and in columns that need 20 to 30 stages. The packing are widely applied in the gas absorption, vacuum processes and pilot scale units (Henley and Seader, 1981). This can be explained, considering that: the packed columns can contain packing made of ceramic or plastic, desirable characteristics required for corrosive systems (very common in gas absorption processes), show characteristics of efficiency and pressure drop, critical factors in the vacuum distillation (often used to separate thermally sensitive mixtures, suffering decomposition and/or polymerization at high temperatures) are cheaper than the tray columns when employed less than 76 cm diameter. Another recommendation, for the preferential use of packed column, is made when you want separate systems with a tendency to form foams, since the tray columns have a higher degree of agitation (Perry and Green, 1999).
According to Perry \& Green (1999), the main restrictions on the use of packed columns are: 1) when multiple feeds and/or multiple side streams and condensers and/or intermediary reboilers are required (tray valves are desirable in these cases); 2 ) when a periodic cleaning must be done inside the column due to certain characteristics of the system to be distilled (trays are easier to clean), 3) when data for the design of packed distillation columns are not available for certain mixtures (projects of tray columns are better established than for packed columns).
None of the three restrictions limit the use of packing to obtain anhydrous ethanol by extractive saline distillation. Doubts could arise with respect to the periodic cleaning of the interior of the column, needed for the alleged deposition of salt. This would not be necessary because the salt would tend strongly to remain in solution. Side devices are not required and the project data can be obtained through studies on a pilot scale, emphasizing the importance of this work.
The packed distillation columns must be fitted with a good distribution of liquid through the interior of the packing, to promote fluid turbulence and mass transfer by liquid dispersion. This allows for a greater contact between the liquid and vapor phases, increasing the efficiency of separation. At low flow rate of steam and/or liquids, or if the feeding of the liquid is not regular distributed over the packing, the liquid will tend to descend the walls of the column, forming preferential channels. Thus, the upward flow of steam is bypassed by the middle column, without a proper contact between the phases. In very small flow rates, the liquid may be insufficient to wet the packing surface. Therefore, it is strongly recommended an adjusted flow condition and the use of distributors, what can improve the wettability of the interior of the column (Henley and Seader, 1981).
The proposal of a generalized correlation for the HETP is a difficult task, because packed columns are equipments of continuous contact, so that the modeling of the phenomenon is more powerful when done by balances in the differential element of the packing. However, the use of HETP in specific situations provides reliable results and, in many cases, is the only possible systematic (Caldas \& Lacerda, 1988).

### 2.2 Part B: performance of structured packing

In the field of distillation, structured packings have been established for several decades. They are preferred where liquid loads are acceptable, a high separation performance is required and low pressure drop is of importance (Fischer et al., 2003).
The first generation of structured packing was brought up in the early forties. In 1953, it was patented a packing named Panapak ${ }^{\text {TM }}$, made of a wavy-form expanded metal sheet, which was not successful may due to maldistribution or lack of good marketing (Kister, 1992).

The second generation came up at the end of 1950's with the highly efficient wire mesh packings, as Goodloe ${ }^{\mathrm{TM}}$, Hyperfil ${ }^{\mathrm{TM}}$ and Koch-Sulzer. Until the 70's, those packings were the most used in vacuum distillation due to their low pressure drop per theoretical stage. However, high cost, low capacity and high sensitivity to solids have prevented the utilization of wire mesh packings, except in vacuum distillation.
The corrugated structured packings, introduced by Sulzer by the end of the 70 's, have initiated the third generation of structured packed columns. High capacity, lower cost, less sensitivity to solids while keeping a high performance, have made them competitive in relation to other column internals. The 1980's have perceived a growing popularity of those packings, especially on revamps in oil and petrochemical plants (Nicolaiewsky, 1999).
Those structured packings, made of corrugated metal sheets, had their surfaces treated, chemical or mechanically, in order to enhance their wettability and, consequently, their wetted area, improving their performance. The way wetted area is created, maintained and renewed, related to different surface geometry, has a remarkable effect not only on packing efficiency, but also on the performance of packed columns (Nicolaiewsky et al., 1999).
Spiegel and Meier (2003) summarized in the Figure 3 the evolution of the structured packings, concluding that no better performance was achieved with various packings of similar geometry. In 1994, a new geometry was developed and called as Optiflow and, in 1999, an improved structure of corrugated sheet packings, the MellapackPlus was developed based on CFD simulations and experimental tests. This new structure, compared with conventional Mellapak, has the pressure drop remarkably lowered and the maximum useful capacity could be extended uo to $50 \%$.


Fig. 3. History of structured packings (Spiegel and Meier, 2003)
According to Shi and Mersman (1985), the effective interfacial area includes not only films on the packing surface but also drops, jets and sprays which flow through the voids of the packed bed. In truth, wetted area can be divided into two parts: one occupied by the liquid film flowing over the surface of the packing and the other, the stagnant liquid. In gas absorption, the fraction of the wetted area occupied by the stagnant liquid soon becomes saturated with gas, and as renewal of that liquid is insignificant; it does not contribute to mass transfer. However, in distillation, these portions of stagnant liquid are also effective in the separation (Puranik and Vogelpohl, 1974).

The first fundamental model for structured packing efficiency is attributed to Bravo et al. (1985), applied to Sulzer gauze packings, in which the effective interfacial area should be considered equal to the nominal packing area. The pressure effect was not included in that model, due to the vacuum conditions on the tests, involving low liquid flow rates and films with lower resistance to mass transfer (Orlando Jr. et al., 2009).
Later, in 1987, Fair and Bravo proposed the following equations to predict the wetted area of corrugated structured packings:

$$
\mathrm{a}_{\mathrm{e}}=\beta \cdot \mathrm{a}_{\mathrm{p}}(34)
$$

where $\beta=0.50+0.0059$ ( $\%$ flood) and $\beta=1.0$ for above $85 \%$ flood. The above equations mean that the effective interfacial area is always lower than the nominal packing area.
Based on measurements of widths of liquid films flowing over inclined surfaces, Shi and Mersmann (1985) have established a correlation for the estimation of wetted area. For the liquid film thickness, they used Nusselt's equation. The authors' correlation for the wetted area took into account the influence of physical properties like viscosity, surface tension and contact angle, with a great influence of the latter. The authors found out that a small variation on contact angle would cause a large influence on the wetted area, which is not reasonable according to findings of Nicolaiewsky et al. (1999), in which work correlations for the estimation of liquid film width and thickness were proposed to be used on a wetted area model in packed columns containing structured packing.
Shi and Mersmann's (1985) correlation for sheet metal structured packings can be written:

$$
\begin{equation*}
\frac{a_{e}}{a_{p}}=F_{S E} \frac{29.12\left(W_{e L} F_{r L}\right)^{0.15} S^{0.359}}{R_{e L}^{0.2} \varepsilon^{0.6}(1-0.93 \cos \theta)(\sin \gamma)^{0.3}} \tag{35}
\end{equation*}
$$

in which $F_{S E}$ accounts for variations in surface enhancements and the contact angle $\theta$ accounts for surface material wettability. For sheet metal packing, the authors stated that:

$$
\cos \theta=0.9 \quad \text { for } \sigma \leq 0.055 \mathrm{~N} / \mathrm{m}
$$

$$
\cos \theta=5.211 \times 10^{-16.835 \sigma} \quad \text { for } \sigma \geq 0.055 \mathrm{~N} / \mathrm{m}
$$

$$
\begin{gather*}
R_{e L}=\frac{4 \delta u_{L e} \rho_{L}}{\mu_{L}}  \tag{36}\\
F_{r L}=\frac{u_{L}^{2}}{S g}
\end{gather*}
$$

$$
\begin{equation*}
W_{e L}=\frac{u_{L}^{2} \rho_{L} S}{\sigma g_{c}} \tag{38}
\end{equation*}
$$

Henriques de Brito and coworkers (1994) measured the effective interfacial area of sheet metal structured packings such as Mellapak 125Y, 250Y and 500Y. Their results have demonstrated that the effective area can be much higher than the packing surface area due to instabilities in liquid flow, such as ripples, waves, detachment of the film into liquid
showers, etc. The resulting correlation for all measurements is a function of the Reynolds number for the liquid phase, as follows:

$$
\begin{equation*}
\frac{a_{e}}{a_{p}}=0.465 R_{e L}^{0.30} \tag{39}
\end{equation*}
$$

It must be pointed out that the authors have not checked the correlation with fluids with different densities and viscosities.
Later on, Rocha et al. $(1993,1996)$ developed a mechanistic model aiming the design and optimization of CSSP (continuous separation structured packing) distillation columns of the metallic corrugated type, also applied to absorption and stripping processes. Liquid holdup prediction was the key to the development of correlations to measure pressure drop, capacity and mass transfer efficiency in the packing. In their model, Rocha and coworkers used Shi and Mersmann's (1985) correlation in order to evaluate the interfacial area available for mass transfer and the liquid holdup present in the packing. Those correlations involved parameters related to surface treatment, as contact angle on the packing surface, as well as packing geometry, liquid and vapor flow rates and physical properties of the system (Orlando Jr. et al., 2009). For the estimation of the liquid side mass transfer coefficient, Rocha et al. (1996) used a correlation developed by Brunazzi and coworkers (1995) for the evaluation of effective areas in absorption columns containing Mellapak 250Y and Sulzer BX.
Rocha et al. (1993) studied correlations to calculate flooding velocity and mass transfer efficiency by using the concept of HETP for distillation columns filled with structured packings. The authors observed that there are few correlations to predict HETP values amd most of them need empirical constants or exponents for their calculation. The disadvantage is that these values are not reported for all the packings and all the sizes available. It was used the Billet (Billet, 1987), Spiegel and Meier (Spiegel \& Meier, 1987) and Bravo et al. (1985) correlations for the HETP calculation and the deviation between them was $11 \%$.

Billingham and Lockett (1999) studied very small modifications to structured packing in order to increase the capacity. It was tested the air-water system and the cryogenic distillation. It was done three bricks of aluminum Flexipac 1Y in the initial experiments and in the other experiments, the packing was removed and the bricks disassembled and repinned together with each alternate sheet staggered in a vertical direction. For the cryogenic distillation, a larger specific surface area packing than Flexipac was used. The authors observed that the key is to reduce the pressure drop associated with vapour entry into the bricks, facilitating the passage of liquid from the bricks. Although the modified packings have increased capacity, HETP is about $25 \%$ higher than that the unmodified packings. To overcome this problem, another packing was used with the bricks having a flat top and a staggered base and were made from sheets of two different lenghs arranged alternatively. The modified packing had about $15 \%$ more capacity and the HETP has the same value of the unmodified packings.
None of those models mentioned so far considered the effect of vapor flow and thus can only be used with low vapor rates. However, since industrial columns often operate above the loading point, it was necessary to develop a correlation for effective interfacial area which was valid for a wide range of vapor rates (Xu et al., 2000). Using Billet and Schultes' model (1993) for effective interfacial area (which is very similar to Shi and Mersmann's model, but at least was validated with experimental data), Xu et al. (2000) introduced in that model the Marangoni effect. The authors considered that the surface tension positive
systems should have a higher interfacial area than the neutral and negative systems because the liquid films are more stable. This result is in agreement with measurements of contact angles formed by liquid films of diverse properties, in flat or textured surfaces (Nicolaiewsky and Fair, 1999), in which it was clear that texturing of surfaces would enhance wetting characteristics of positive systems. Xu et al's model (2000) was validated for three structured packing (Gempak 2.5A, AW7 and AW12), with three test systems (methanol-isopropanol, water-acetic acid and methanol-water), at two operating pressures ( 710 and 260 mmHg ), having achieved an average $10.2 \%$ deviation of model prediction. The deviations are $20 \%$ approximately for $90 \%$ of the data points.
Olujic et al. (2003) made experimental studies in a distillation column at total reflux to report the capacity of a new generation of Montz structured packing. According to the authors, the major feature of Montz B1 is a smooth bend in the bottom third of the corrugation with continuously increasing corrugation base width. The experiments were done using a bed height of approximately 3.3 m and the system cyclohexane/heptanes was utilized, under different conditions of pressure, $0.17,0.33,1.03$ and 4.14 bar. Five different dimensions of structured packing were investigated with $a_{p}\left(\mathrm{~m}^{2} / \mathrm{m}^{3}\right)$ of $244,247,250,346$ and 350 . The number of equilibrium stages was calculated from the distillate and bottom compositions using the Fenske equation and an average relative volatility. Among the various results published by the authors, Figure 4 shows the dimensionless product of the specific surface area and HETP as a function of the operational pressure for both packing sizes. The figure concludes that larger surface area packing appear to use their available surface less efficiently. The performance enhancement in case of 350 series packings is $21 \%$ with respect to the standard size. The relatively high efficiency is similar as the original packing in its range of application. Because of that characteristic, columns equipped with B1-250 packing can be revamped with B1-250M packing.


Fig. 4. Relative surface utilization efficiency as a function of operating pressure (total reflux)
In 2006, results from continuous feed and total reflux distillation experiments, carried out with a common type and size structured packing using two- and three-component mixtures of common alcohols and water, were published (Mori et al., 2006). The composition profiles
measured with the three component mixture were used to validate the rate-based (nonequilibrium) model developed at the Nagoya Institute of Technology (NIT), which appeared to be highly accurate, but also sensitive to the choice of the predictive method for the interfacial area. The rate-based, non-equilibrium (NEQ) approach adopted at NIT (Mori et al., 1996, 1999, 2002) includes Bravo et al. (1985) correlations for vapor and liquid side mass transfer coefficients that differ to some extent from those employed in the Delft model (Olujic et al., 1999). Two simple empirical models for effective specific area were considered in this work. First one is that introduced by Olujic and coworkers (1999) for Montz B1-250 packing and the other one is proposed by Henriques de Brito et al. (1994), for Mellapak packings. The authors concluded that the Delft model overpredicts the measurements at higher F factors to such an extent that it may be considered as safe or conservative. The NEQ model developed at NIT, in conjunction with Henriques de Brito et al. (1994) correlation for effective area, proved to be capable of reproducing all the measured composition profiles very well, regardless of the water content of the feed.
Also, in 2006, Mori et al. presented results of continuous feed and total reflux distillation experiments carried out with a common type and size structured packing, Montz-pack B1250, using two (methanol-water) and three (methanol/ethanol/water) component mixtures of common alcohols and water. The packing is a conventional corrugated sheet metal structured packing with a regular shallow embossed unperforated surface, a corrugation inclination angle of $45^{\circ}$ and an element height of 0.196 m . The experiments were done in a 210 mm diameter distillation column, with a total packed height of 2.156 m and the packed bed consisted of four sections. The experimental packing efficiency, expressed as HETP, under total reflux conditions, was calculated using the Fenske equation to estimate the number of equilibrium stages. The rate-based, non-equilibrium (NEQ) model was used because it does not require any empirically determined packing specific constant, just the main dimensions of corrugated sheets as well as the corrugation inclination angle. The model includes Bravo et al. (1985) correlations for vapor and liquid side mass transfer coefficients that differ from the Delft model. Two simple empirical model for the calculation of the effective specific area was considered: the first one, by Olujic et al. (1999) for B1-250 packing (equation 40) and the other one was proposed by Brito et al. (1994) for Mellapack packings (equation 41).

$$
\begin{gather*}
\frac{a_{e}}{a_{p}}=\frac{1}{1+\left(0.000002143 u^{-1.5}{ }_{L S}\right)}  \tag{40}\\
\frac{a_{e}}{a_{p}}=0.465 \operatorname{Re}^{0.3}{ }_{L} \tag{41}
\end{gather*}
$$

Equation 40 was originally developed for Mellapak and is assumed here to be valid for other similar sheet metal packings, including Montzpak B1-250. The results, at total reflux conditions, is presented in Figure 5, that shows the mass transfer efficiency (the average HETP of B1-250) as a function of vapor load factor (F-factor). In the figure, Feed 1 refers to a low water content and the Feed 2 to a feed with relatively high water content. In both cases, the efficiency slowly decreases with increasing F-factor. According to the authors, a pronounced trend was observed with the B1-250 packing because is it an inherent characteristic of the Delft method (Olujic et al., 1999 cited by Mori et al., 2006).


Fig. 5. Comparison of calculated and measured packing efficiencies at total reflux conditions
Ceramic foam packing has been known for many years and has a wide range of applications due to its low density and attractive thermal, mechanical, electrical, and acoustical properties. In a recent paper (Lévêque et al., 2009), its performance was evaluated as a distillation packing material. The hydraulic characteristics of the foam were experimentally determined for gas-liquid countercurrent flow using an air-water system. The performance in terms of pressure drop per unit height and flooding behavior was quite low compared with classical distillation packing materials (Sulzer M250Y, Sulzer CY and Pall rings). The liquid hold-up of the foam packing increased with increased liquid-gas loading in the loading zone, and the liquid hold-up was greater than other classical packing materials. Mass transfer efficiency was determined over the entire operating range using a cyclohexane/ $n$-heptane system at atmospheric pressure under total reflux. The foam packing performance was very good, with a HETP of 0.2 m and increasing mass transfer with increasing gas and liquid superficial velocities inside the packing (Lévêque et al., 2009). Last year, a French group from Université de Toulouse (Bessou et al., 2010), sponsored by Sulzer and Snecma Propulsion Solide, has developed a new structured packing, made of carbon, named The Sepcarb 4D packing, presenting its performance characteristics. The advantages of the packing rest on being inert, corrosion-resistant; has very low density ( 40 $\left.\mathrm{kg} / \mathrm{m}^{3}\right)$, with tubes of small thickness $(0.2 \mathrm{~mm})$. The separation efficiency has been determined using the HETP concept on distillation experiments with cyclohexane/nheptane system at atmospheric pressure and total reflux. The best results were obtained with wall wipers, which improved liquid redistribution between packing cylinders and involved low wall effects. HETP calculated was 0.2 m , which corresponds to a good transfer performance when compared to classical packings such as Mellapak 250Y, Mellapak 425Y and Pall rings.
Performance of a distillation column, operating continuously with a mixture of known composition $\left(\mathrm{C}_{8}-\mathrm{C}_{14}\right)$, containing Sulzer DX SS structured packing, has been evaluated. Prior to the experimental tests, simulation studies using commercial software $\mathrm{PRO} / \mathrm{II}$ ® were performed in order to establish the optimum operational conditions for the distillation, especially concerning operating pressure, top and bottom temperatures, feed location and reflux ratio. The results of $\mathrm{PRO} / \mathrm{II}$ ® were very similar to the analysis of the products obtained during continuous operation, therefore permitting the use of the properties
calculated by that software on the theoretical models investigated. Five theoretical models available in the literature (Bravo, Rocha and Fair, 1985; Rocha, Bravo and Fair, 1993, 1996; Brunazzi and Pagliant, 1997; Carlo, Olujić and Pagliant, 2006; Olujić et al., 2004) and an empirical model (Carrillo and coworkers, 2000) have been compared. Modifications concerning calculation of specific areas were performed on the correlations in order to fit them for gauze packing HETP evaluation. As the laboratory distillation column was operated continuously, different HETP values were found by the models investigated for each section of the column. The low liquid flow rates in the top section of the column are a source of error for HETP evaluation by the models; therefore, more reliable HETP values were found in the bottom section, in which liquid flow rates were much greater. Among the theoretical models, Olujić et al. (2004) has shown good results relatively to the experimental tests. In addition, the former model by Bravo, Rocha and Fair (1985) underestimates HETP values; however, with the modifications proposed in this work, it has achieved more realistic performance prediction, remaining a good choice for gauze packing HETP evaluation. Having the advantage of avoiding the calculation of effective area and mass transfer coefficients, an empirical model proposed by Carrillo and coworkers (2000) was also investigated, showing low deviations compared to the theoretical models tested.
Among the short-cut methods for the estimation of column efficiency, Carrillo and coworkers (2000) have proposed a modification of the Lockett equation (1998) to be used for HETP estimation of Sulzer BX packing. The correlation was proposed to be a function of the gas flow factor, densities of the liquid and vapor phases and the system pressure. The HETP values calculated by the modified equation have shown a good fit, compared to the published experimental data available.
Later, Carlo, Olujić and Pagliant (2006) used the absorption column studies developed by Brunazzi and Pagliant (1997), and made some modifications on the liquid side mass transfer coefficient, to adapt those correlations for HETP evaluation of distillation columns. Olujić's model (1997) was developed to predict hydraulic and separation performance of corrugated sheet structured packing in distillation systems. Since 1997, until its last version (Olujić et al. (2004), the model, named the Delft model, has been enhanced through tests using Montz B1 and BSH packings. A complete evaluation of Delft's model has been accomplished by Fair and coworkers (2000), showing that it overestimates the effective superficial area for structured packing column design. In order to compensate for that deviation, Olujić et al. (2004) have adapted Onda's correlation (1968) apud Olujić et al. (2004) to be used with structured packing columns.
In 2009, two works deal with the HETP evaluation using distinct base lube oil mixtures in a lab-scale distillation column, of 40 mm of nominal diameter, having 4 sections of 550 mm each containing Sulzer DX (gauze) structured packing, as the contacting device (Machado et al. and Orlando Jr. et al., 2009). Orlando Jr. et al. (2009) made several tests with a hydrocarbon mixture $\left(\mathrm{C}_{8}-\mathrm{C}_{14}\right)$ to evaluate HETP using six HETP correlations to find out which is the most appropriate for structured packed columns with medium distillates. The theoretical models investigated are: Bravo et al. (1985), Rocha et al. $(1993,1996)$, Brunazzi and Pagliant (1997), Carlo et al. (2006) and Olujic et al. (2004). As the laboratory distillation column was operated continuously, different HETP values were found by the models for each section of the column. Olujic et al. (2004) was the best method, showing good results, together with the correlation of Carrillo et al. (2000), in which low deviations were obtained. The average deviation varied from 8 to $47 \%$. The deviations can be explained by the fact that
most of the models have been proposed from testes using corrugated sheet structured packing elements, which performs differently from gauze type packing.
In the work of Machado and coworkers (2009), the authors worked with two mixtures, a heavier one composed of neutral medium and bright stock and another composed of spindle and neutral light. Simulation studies using the PRO II software had been performed in order to establish the best operating conditions in the distillation unit. Concerning the empirical models, a comparison between the Lockett (1998) and Carrillo et al. (2000) models was done. Among the theoretical models, Olujic et al. (2004) was chosen for being one of the most recent and robust. According to the authors, unfortunately, neither mass transfer model was able to properly describe the base lube oil distillation. Olujic et al. (2004) model yielded underestimated area values, by using Onda's correlation (Onda et al., 1968), but a modified version proposed by Orlando Jr. et al. (2009) provided more realistic values for the effective areas. It was concluded that the nature of the mixtures had no influence on HETP deviations, pointing out that the low vapor flowrate inside the column was the most influential variable. Large deviations varying from 27 to $70 \%$ were obtained for all the mixtures and using all the methods.
Finally, Li et al. (2010) used a special high-performance structured packing, PACK- ${ }^{13} \mathrm{C}$, with a surface area of $1135 \mathrm{~m}^{2} / \mathrm{m}^{3}$ and the first stable isotope pilot-scale plant using structured packing was designed. The height and inner diameter of the distillation column were 20 m and 45 mm , respectively, and the height of the packing bed was 18 m . The raw materials utilized were only high purity CO gas and liquid notrogen. When the F-factor changes from 0.18 to $0.90 \mathrm{~m} / \mathrm{s}$, the number of theoretical plates per meter decreases from 30 to 20 . The new structured packing was a combination of the advantages of structured and random packing, as showed in Figure 6. The inclination angle was $45^{\circ}$, the height of corrugation was 2.5 mm , the porosity was 0.77 and the silk diameter was 0.085 mm .


Fig. 6. Illustration of the structured packing PACK-13C (Li et al., 2010)
The minimum theoretical plates, at total reflux, was calculated by the Fenske equation. The authors concluded that the PACK-13C structured packing exhibits very high performance in isotope separation, combining the advantages of high theoretical plate numbers of the random packing and the excellent hydrodynamic properties of structured packing. Figure 7 confirms the characteristics of the structured packing relating the theoretical plates number and factor F .


Fig. 7. The theoretical plates of PACK-13C (Li et al., 2010)

## 3. Conclusions

The use of packed columns for continuous contacting of vapors and liquids is well established in the chemical industry, nowadays. The design of the columns require a knowledge of the height of a transfer unit and this chapter had as main objective the description of the present correlations, relating their advantages and disadvantages, for random and structured packing.
Among the researches encountered in the literature and cited in this chapter, it is important to have a model that describes the fluid dynamic relationships in packed columns with countercurrent flow of the gas and liquid phases to describe up the flood point. It is so important because above this point, the liquid accumulates to such an extent that column instability occurs. The disadvantage of some correlations relies on the fact that many parameter characteristics is only obtained graphically, what introduces deviations in the calculation of areas and HETP.
For the distillation in packed columns, it was ascertained that the resistance in both phases, liquid and vapor phases, should be taken into account in the HETP evaluation.
About the packing, new random and structured packing have been studied, but the difficulty in HETP representation remains the problem, due to the fact that it is so difficult to find a correlation that covers all systems with different physical properties and different nominal sizes of the packing.
Moreover, normally, HETP is substantially constant over a wide range of vapor flows; on the other hand, vapor flow varies increasing or decreasing the mass transfer depending on the liquid phase. Because of that, HETP is not constant along the column and it is convenient to define one value that which may be used for design purposes. Due to these factors, the correlations proposed, empirical or theoretical, do not reach the real value of HETP for any system studied.

Finally, to better evaluate HETP, it is also important to choose a thermodynamic model that can represent the behavior of the liquid-vapor equilibrium and complex methodologies to calculate the theoretical number of stages.

## 4. Nomenclature

$d_{P}$ - nominal size of the packing
$\lambda$ - inclination ratio between the equilibrium and operation straight
$\beta$ - fraction of surface used for mass transfer
$P$ - pressure of the system
$G$ - mass flow of the phase
$\mathrm{G}_{\mathrm{L}}$ - liquid mass flow
$G_{V}$ - vapor mass flow
G - V - gas or vapor rate
L - liquid rate
M - molecular mass
$\mathrm{M}_{\mathrm{L}}$ - molecular weight of liquid
$\mathrm{M}_{\mathrm{v}}$ - molecular weight of vapor
$\rho$ - density
$H$ - height representation of the mass transfer unit
$\mathrm{H}_{\mathrm{G}}-\mathrm{H}_{\mathrm{V}}$ - height of gas-side phase transfer unit (m)
$\mathrm{H}_{\mathrm{L}}$ - height of liquid-side phase transfer unit (m)
$\mathrm{H}_{\mathrm{OG}}-\mathrm{H}_{\mathrm{Ov}}$ - height of an overall gas phase transfer unit (m)
$g_{C}$ - conversion factor between strengh and mass
$C a_{L}$ - capilar number
$\mathrm{C}_{\mathrm{fL}}$ - coefficient for effect of approach of flood point on liquid-phase mass transfer
$\mathrm{C}_{\mathrm{pk}}$ - packing characteristic
h - operating holdup $-\mathrm{m}^{3} / \mathrm{m}^{3}$
c - void fraction
a - corrugation angle
$\gamma$ - angle with the horizontal for falling film or corrugation channel
$\Theta$ - contact angle
g - gravity acceleration
R - universal constant of the gases
T-absolute temperature
$S$ - side dimension of corrugation - $m$
$u_{L s}$ - liquid phase superficial velocity $-\mathrm{m} / \mathrm{s}$
$u_{G s}$ - vapor phase superficial velocity $-\mathrm{m} / \mathrm{s}$
$\mu_{L}-$ liquid viscosity $-\mathrm{kg} / \mathrm{m} . \mathrm{s}$
$\mu_{v}-\mu_{G}-$ vapor viscosity $-\mathrm{kg} / \mathrm{m} . \mathrm{s}$
$\mu_{w}$ - water viscosity - $\mathrm{kg} / \mathrm{m} . \mathrm{s}$
$\rho_{L}$ - liquid density $-\mathrm{kg} / \mathrm{m}^{3}$
$\rho_{v}$ - vapor density $-\mathrm{kg} / \mathrm{m}^{3}$
$\rho_{w}$ - water density - $\mathrm{kg} / \mathrm{m}^{3}$
$\varepsilon-$ void fraction of packing
$\Phi$ - packing parameter (function of packing type, size and $\mathrm{G}_{\mathrm{L}}$ )
$\psi$ - ratio (density of water/density of liquid)
$d^{\prime}{ }_{p}$ - diameter of a sphere with the same superficial area of the packing element
dc - column diameter
$S_{\text {C }}$ - Schmidt number
$S_{C V}$-Schmidt number of the vapor phase
$\mathrm{S}_{\mathrm{CL}}$ - Schmidt number of the liquid phase
$D$ - diffusivity
$D_{L}-$ liquid diffusion coefficient $-\mathrm{m}^{2} / \mathrm{s}$
$\mathrm{D}_{\mathrm{V}}$ - vapor diffusion coefficient $-\mathrm{m}^{2} / \mathrm{s}$
$\sigma$ - liquid surface tension $-\mathrm{N} / \mathrm{m}$
$\sigma_{c}-$ critical surface tension $-N / m$
Z - height of the packed bed
N - number of theoretical stages
m - slope of equilibrium line
$\mathrm{a}_{\mathrm{e}}$ - effective interfacial area ( $\mathrm{m}^{2} / \mathrm{m}^{3}$ )
$\mathrm{a}_{\mathrm{w}}$ - wetted surface area of packing $\left(\mathrm{m}^{2} / \mathrm{m}^{3}\right)$
$a_{p}$ - specific surface of the packing $\left(\mathrm{m}^{2} / \mathrm{m}^{3}\right)$
$\mathrm{k}_{\mathrm{G}}-\mathrm{k}_{\mathrm{V}}$ - gas-phase mass transfer coefficient
$\mathrm{k}_{\mathrm{L}}$ - liquid-phase mass transfer coefficient
$\mu_{r}$ - relation between liquid viscosity at the packing bed temperature and viscosity of the water at reference temperature of $20^{\circ} \mathrm{C}$
$R_{e L}=\frac{4 \delta u_{L e} \rho_{L}}{\mu_{L}}$
(Reynolds number for liquid)
$F_{r L}=\frac{u_{L}^{2}}{S g} \quad$ (Froude number for liquid)
$W_{e L}=\frac{u_{L}^{2} \rho_{L} S}{\sigma g_{c}} \quad$ (Weber number for liquid)

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## Mass Transfer in Chemical Engineering Processes

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This book offers several solutions or approaches in solving mass transfer problems for different practical chemical engineering applications：measurements of the diffusion coefficients，estimation of the mass transfer coefficients，mass transfer limitation in separation processes like drying，extractions，absorption，membrane processes，mass transfer in the microbial fuel cell design，and problems of the mass transfer coupled with the heterogeneous combustion．I believe this book can provide its readers with interesting ideas and inspirations or direct solutions of their particular problems．

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