STUDIES CONCERNING MASS AND HEAT TRANSFER ON B7 STRUCTURED PACKING

by

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The paper presents theoretical and experimental data concerning mass and heat transfer on B7 structured packing, used for deuterium separation in distillation column. The first section of the paper is dedicated to mass transfer study for hydrogen distillation, and the second section to mass and heat transfer for water distillation. Mathematical model verification was carried out with experimental data, obtained from two laboratory distillation plants for deuterium separation. The experimental data concerning B7 ordered packing efficiency for hydrogen cryogenic distillation at about –250 °C level were obtained from the first plant, and the second plant provided data concerning mass and heat transfer on the same packing for deuterium separation by water vacuum distillation at about 60 °C level. Height of transfer unit and mass and heat transfer coefficients, evaluated theoretically and experimentally, are comparable with those from chemical industry separation processes. This fact justifies the use of multi-tubular column model for transfer process description at distillation column equipped with B7 structured packing.

Key words: isotopic distillation, structured packing, deuterium

INTRODUCTION

Isotope separation by distillation represents an important area of nuclear energetics. This concerns heavy water production and heavy water detritiation. The distillation columns are frequently equipped with structured packing. The packing provides a large contact surface and introduces low pressure drop. It is used on large scale in chemical industry for mixture separation by distillation, absorption, desorbtion, *etc*.

To design or simulate the functioning of distillation columns, it is necessary to provide quantities which characterize the behavior of liquid-vapor contact elements, such as the height of transfer unit or the mass and heat transfer coefficients.

The paper presents the results of theoretical and experimental researches effected to describe mass and heat transfer on B7 structured packig on hydrogen isotope separation by distillation. This packing is produced by National Research and Development Institute for Cryogenics and Isotopic Technologies – ICIS Rm. Valcea, Romania. The first time B7 packing was manufactured to equip distillation columns of the Romanian heavy water plants. Firms Sulzer and Koch-Glitsch are acknowledged producers of structured packing.

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MASS TRANSFER STUDIES FOR HYDROGEN DISTILLATION

The liquid flow is pellicular at distillation column equipped with structured packing, presented in fig. 1.

If the void space of packing is divided in parallel tubes, with height equal to packing bed height, a multi-tubular column is obtained. Each tube simulates distillation in a wetted-wall column [1, 2, 3].

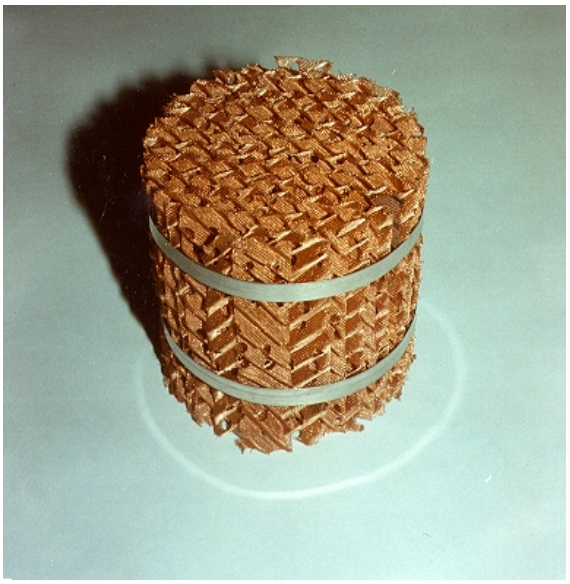

Figure 1. B7 structured packing

Mathematical model

A measure of liquid-vapor contact element efficiency is the height of the transfer unit *HTU*, based on the two film theory. It may be theoretically evaluated using the following relations [1]:

$$(HTU)_{OV} \quad \frac{V\, d_{ech}}{D_c^2\, \varepsilon\, \pi\, K_{OV}} \tag{1}$$

$$\frac{1}{K_{OV}} \quad \frac{1}{k_V\, P} \quad \frac{m}{k_L\, \rho_m} \tag{2}$$

where:

V – vapor flow rate in column, [mol h^{-1}],

d_{ech} – packing equivalent diameter, [m],

D_c – column diameter, [m],

ε – void space of packing, [m^3 m^{-3}],

K_{OV} – overall (OV) mass transfer coefficient related to the vapor phase, [mol m^{-2} h^{-1}],

k_V – partial vapor film mass transfer coefficient, [mol m^{-2} h^{-1} atm^{-1}], where 1 atm = 101.325 kPa,

k_L – partial liquid film mass transfer coefficient, [m h^{-1}],

m – slope of the equilibrium curve,

P – total pressure (absolute), [atm], and

ρ_m – molar density of liquid, [mol m^{-3}].

The mass transfer coefficient in vapor phase was calculated with Johnstone and Pigford relation, eq. (3), and in liquid phase with V. G. Levici correlation for Sherwood number, eq. (4) [2, 3].

$$(k_V\, P)\frac{A}{V} \quad \frac{\eta_V}{\rho_V\, D_V}^{2/3} \quad 0.033\mathrm{Re}_V^{0.23} \tag{3}$$

$$k_L \quad \frac{\mathrm{Sh}_L\, D_L}{\delta_L^c} \quad \frac{c\,\mathrm{Re}_L^{1/2}\,\mathrm{Sc}_L^{1/2}\, D_L}{\delta_L^c} \tag{4}$$

where:

A – area of free column section, [m^2],

η_V – vapor viscosity, [kg m^{-1} h^{-1}]

ρ_V – vapor density, [kg m^{-3}],

D_V – molecular diffusion coefficient in vapor phase, [m^2 h^{-1}],

D_L – molecular diffusion coefficient in liquid phase, [m^2 h^{-1}]

Re – Reynolds number,

Sh – Sherwood number,

Sc – Schmidt number, and

δ_L^c – thickness of the mass transfer boundary layer of the liquid phase [m].

For the wetted wall column the "c" constant is equal to 1/2.

Verification of the mathematical model proposed for *HTU* theoretical evaluation is carried out by comparison with *HTU* values calculated from the experimental data, *HTU* being the ratio between the height of packing bed (Z) and number of transfer units (NTU):

$$(HTU)_{OV}^{\exp} \quad \frac{Z}{(NTU)_{OV}} \tag{5}$$

If the column is operated at total reflux and the separation factor α is considered constant in the range $y_1 - y_2$, $(NTU)_{OV}$ may be obtained with the Colburn formula [4]:

$$(NTU)_{OV} \quad \frac{2.303}{\alpha \quad 1} \lg \frac{(1 \quad y_1)\, y_2}{(1 \quad y_2)\, y_1}$$

$$2.3 \lg \frac{1 \quad y_1}{1 \quad y_2} \tag{6}$$

where:

y – vapor phase concentration [mole fraction], and

α – separation factor.

Experiments

The experimental plant, by whose means the B7 type ordered packing efficiency, expressed by *HTU*, has been determined is represented schematically in fig. 2 [1]. The principal components of the plant are a condenser-column-boiler ensemble (2-3-5), hydrogen liquefier (1), power supplies for the electrical heaters (8, 9), and measurement and control equipment. The isotopic distillation column for H_2-HD mixture has the inner diameter of 27 mm and at the height of 550 mm it is equipped with the B7 type structured packing. This packing is manufactured of phosphorous bronze weave and it has the following properties: specific surface of 670 m^2/m^3 and void

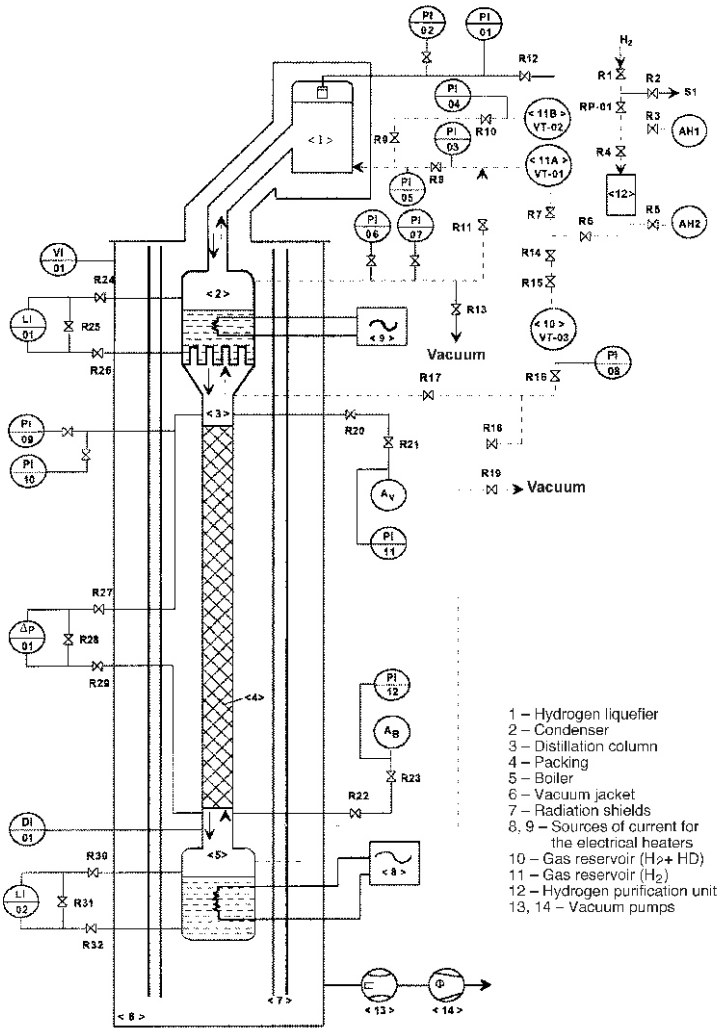

Figure 2. Schematic diagram of the experimental cryogenic distillation plant

space of 95%. The necessary vapor flow rate is provided by a boiler which is equipped with an electrical heater. The condenser attached to the distillation column is multi-tubular and it is cooled with liquid hydrogen. The liquid hydrogen of condenser is for 0.19-0.5 degree colder than the vapors from the column top. This difference permits vapor condensation, so that the liquid reflux is ensured. The vacuum jacket (6) and radiation shields (7) ensure the thermal insulating of the experimental plant cryogenic zone. For the reduction of the entered heat by conduction in residual gas to minimum, the pressure in the vacuum jacked is maintained at 10^{-5}-10^{-6} torr (1 torr = =101.325/760 kPa), by means of the vacuum pumps (13) and (14).

Results

The experiments have been performed at 2, 3 and 4 bar (1 bar = 100 kPa). The parameters measured at ~3 bar are presented in tab. 1, as well as experimentally determined $(HTU)_{OV}$.

The $(HTU)^{exp}$ values were calculated with relations (5), and (6). These values are also presented in fig. 3, beside calculated values.

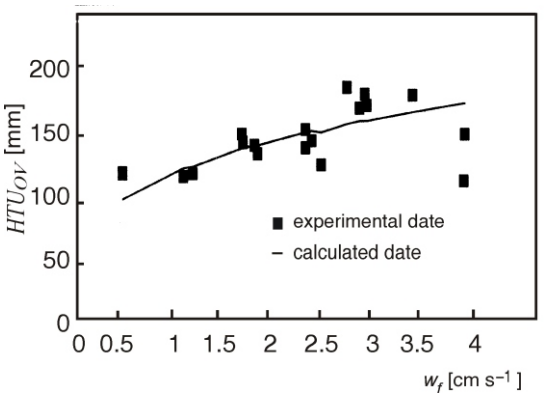

Figure 3. *HTU* variation with the vapour velocity w_f P = 3 bar, T = 24.68 K

Table 1. Hydrogen distillation experimental data

No.	P_{col} [bar]	T_{col} [K]	P_{cond} [bar]	T_{cond} [K]	$y_1\ 10^6$	$y_2\ 10^6$	w_f [cm s^{-1}]	$(HTU)_{OV}^{exp}$ [mm]
1	3.01	24.7	2.88	22.7	88	703	0.53	132.4
2	3.01	24.7	2.91	24.53	93.7	759	0.53	131.3
3	3.07	24.79	2.95	24.6	89.5	742	1.13	128.9
4	2.82	24.4	2.72	24.24	82	696	1.22	131.5
5	3.02	24.71	2.86	24.46	126.2	713	1.7	158.4
6	3.01	24.7	2.86	24.46	119	718	1.71	152.8
7	2.98	24.65	2.8	24.37	88.6	550	1.82	151
8	3	24.68	2.8	24.37	84	563	1.85	144.6
9	3	24.68	2.85	24.4	131	723	2.33	161
10	3	24.68	2.85	24.45	115.5	733.6	2.33	148.8
11	3	24.68	2.8	24.37	89.9	539	2.39	153.6
12	2.78	24.33	2.6	24.04	70.9	558.2	2.48	137
13	2.98	24.65	2.78	24.33	133	563.5	2.74	191
14	3.04	24.74	2.85	24.45	150	708.5	2.86	176.3
15	2.98	24.65	2.81	24.38	168	740.7	2.91	185.8
16	2.96	24.62	2.8	24.37	152	718.5	2.93	177.9
17	3	24.68	2.8	24.37	160	710.8	3.38	184.4
18	2.97	24.63	1.69	22.25	56	508.9	3.88	125.1
19	3	24.68	2.78	24.33	117	670.8	3.89	157.5

P_{col} – pressure in the column, T_{col} – temperature in the column, P_{cond} – pressure in the condenser, T_{cond} – temperature in the condenser, y_1 – deuterium concentration at the column top (mole fraction), y_2 – deuterium concentration at the column bottom (mole fraction), w_f – fictitious vapor speed, *HTU* – height of the transfer unit

MASS AND HEAT TRANSFER STUDIES FOR WATER DISTILLATION

A more complex study concerning mass and heat transfer was conducted at the water isotopic distillation column equipped with B7 structured packing, using the multi-tubular column model. Mass and heat transfer processes were studied by means of the transfer coefficients [5].

Mathematical model

Theoretical evaluation of the packing performance is carried out using previously defined relations for mass transfer (2, 3, 4) and relations for heat transfer. When there is a direct contact between fluids, the overall heat transfer coefficient becomes:

$$K_t \quad \frac{1}{\frac{1}{\alpha_L} \quad \frac{1}{\alpha_V}} \tag{7}$$

Chilton-Colburn analogy was used to express the vapor phase heat transfer coefficient in terms of the vapor phase mass transfer coefficient [6]

$$\alpha_V \quad C_{P_V}\, k_V\, \mathrm{Le}_V^{\,2/3} \tag{8}$$

For the liquid phase, a correlation based on a penetration theory mechanism was used [6]

$$\alpha_L \quad C_{P_L}\, k_L\, \mathrm{Le}_L^{\,1/2} \tag{9}$$

For mass and heat transfer coefficient determination from the experimental data, the following relations were used

$$\frac{\mathrm{d}L}{\mathrm{d}z} \quad \frac{\mathrm{d}V}{\mathrm{d}z} \tag{10}$$

$$\frac{1}{A_L}\,\frac{\mathrm{d}(Lx)}{\mathrm{d}z} \quad \frac{1}{A_V}\,\frac{\mathrm{d}(V\,y)}{\mathrm{d}z}\,\frac{A_V}{A_L}$$

$$= K_y\, a_{1\ v}\,(y \quad y^*) \quad K_y^{'}(y \quad y^*) \tag{11}$$

$$\frac{1}{A_L}\,c_{p_L}\,\frac{\mathrm{d}(Lt_L)}{\mathrm{d}z} \quad \frac{1}{A_L}\,c_{p_V}\,\frac{\mathrm{d}(V\,t_V)}{\mathrm{d}z}$$

$$K_t\, a'_{L\ V}\,(t_V \quad t_L) \quad \frac{\mathrm{d}L}{\mathrm{d}z}\,c_{p_L}\,t_L$$

$$K_t^{'}(t_V \quad t_L) \quad \frac{\mathrm{d}L}{\mathrm{d}z}\,c_{p_L}\,t_L \tag{12}$$

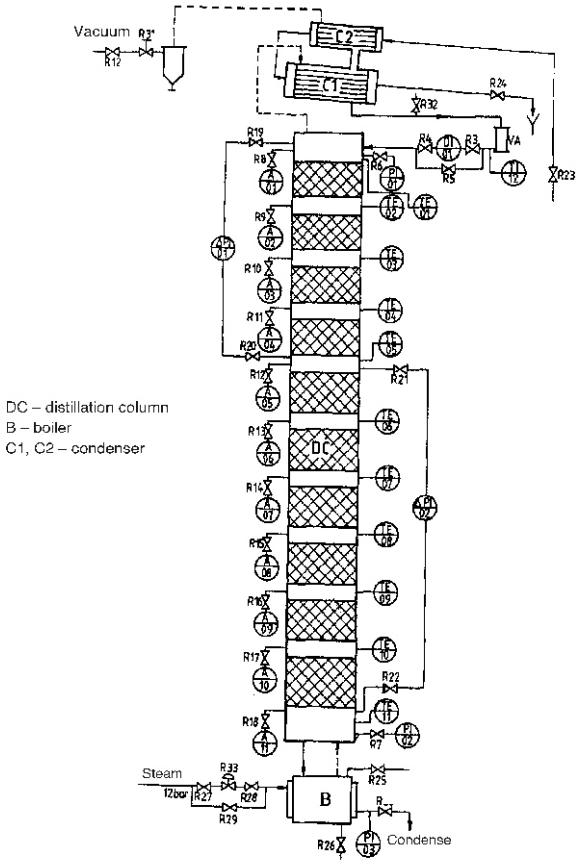

Figure 4. Schematic diagram of the experimental water distillation plant

where:

K_y – mass transfer coefficient, [kmol m^{-2} s^{-1}],
K_t – heat transfer coefficient, [kW m^{-2} °C^{-1}],
α_L – heat transfer coefficient for liquid, [kW m^{-2} °C^{-1}],
α_V – heat transfer coefficient for vapor, [kW m^{-2} °C^{-1}],
c_{p_L} – heat capacity of liquid, [kJ kmol^{-1} °C^{-1}],
c_{p_V} – heat capacity of vapor, [kJ kmol^{-1} °C^{-1}],
Le_L – Lewis number for liquid phase,
Le_V – Lewis number for vapor phase,
x – liquid phase concentration, [mole fraction],
y – vapor phase concentration, [mole fraction],
z – height of packing, [m],
A_L – liquid flow section, [m^2],
A_V – vapor flow section, [m^2],
a_{1-v} – mass transfer interface area, [m^2 m^{-3}],
$a'_{1\ v}$ – heat transfer interface area, [m^2 m^{-3}],
L – liquid flow rate in column, [kmol s^{-1}],
V – vapor flow rate in column, [kmol s^{-1}],
t_L – liquid temperature, [°C], and
t_V – vapor temperature, [°C].

Experiments

The experimental plant for water distillation is represented schematically in fig. 4 [5].

The distillation column has the inner diameter of 108 mm and at 14 m height it is equipped with the B7 type structured packing. Two multi-tubular condensers cooled with water and a film boiler heated with steam provide the column with liquid and vapor. The experimental plant has 11 points for temperature and pressure measurement at the distinct outlets of the column and one point for reflux temperature measurement. The column is insulated with mineral cotton.

Results

The ten percent deutereted water was used for the experiments. The initial value maintains the concentrations in the measurement range of the analyzer, 0.5-99% D/(D+H). The pressure at the column top was maintained at about 100 mm Hg. The column was operated at a vapour capacity factor of 1.4 m^{-2} kg s^{-1}. This factor is defined as $f \quad w\rho^{1/2}$, where w is fictitious vapour

Table 2. Water isotopic distillation experimental data

z [m]	x [% mol]	t [°C]	P_{top} [mm Hg]	ΔP [mm H_2O]	K'_y [kmol m^{-3} s^{-1}]	K'_t [kW m^{-3} °C^{-1}]	$(HTU)_{OV}^{\exp}$ [m]
0.0	0.59	53.3	647.5				
1.3	1.11	53.3			0.132	3.24	0.176
2.6	1.95	55.1		57.75	0.189	2.76	0.098
3.9	4.00	57.4			0.273	4.75	0.063
5.2	6.38	61.2	589.75		0.161	11.7	0.103
6.7	10.21	61.4			0.137	–1.78	0.115
8.1	15.81	63.85			0.159	1.43	0.967
9.5	29.52	62.8		15	0.243	1.25	0.062
10.9	41.76	63.8			0.133	2.06	0.110
12.3	51.46	62.2			0.0673	5.25	0.208
14.2	72.55	65.8	574.75		2.14	–0.0898	0.063

z – height of the packing, x – deuterium concentration in liquid phase, t – vapor temperature, P_{top} – vacuum at the column top, ΔP – pressure drop, K_y – mass transfer coefficient, K_t – heat transfer coefficient, HTU – height of the transfer unit

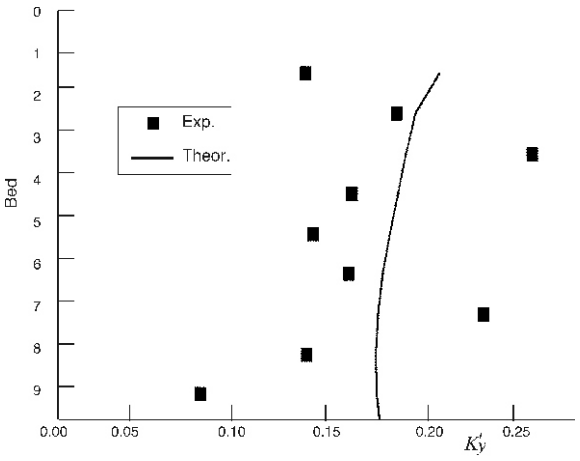

Figure 5. Mass transfer coefficient, K_y' [kmol m^{-3} s^{-1})]

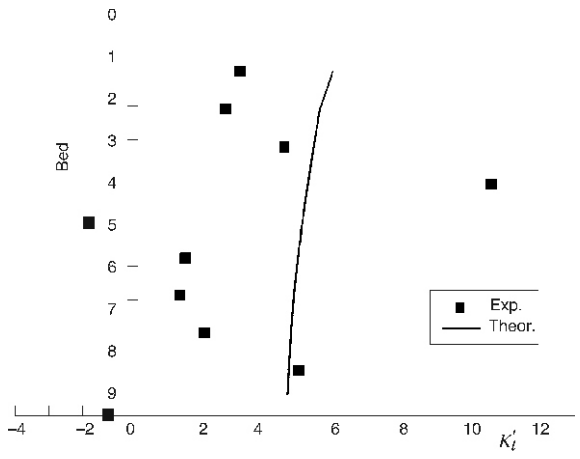

Figure 6. Heat transfer coefficient K_t' [kW m^{-3} °C^{-1}]

speed [m s^{-1}] and ρ is the vapour density [kg m^{-3}]. The experimental data are shown in tab. 2.

These values permit local mass and heat transfer coefficient determination, using relations (10), (11) and (12). The coefficients determined from the experimental data are presented beside the theoretical coefficients in fig. 5 and 6.

DISCUSSION

The experiments for B7 structured packing *HTU* determination were carried out at two temperature levels, about –250 °C for hydrogen isotopic distillation and about +60 °C for isotopic distillation of water. The working pressure and maximum gas rate were ~3 bar and 4 cm/s for hydrogen distillation and ~100 mm Hg and 5 m/s for water distillation. Under those conditions, the *HTU* value range was 0.1-0.2 m. Mass and heat transfer processes were studied on water distillation, the heat transfer coefficient values being between 2 and 12 kW/m°C. The study also included theoretical evaluation of *HTU*. The range of differences between experimental and theoretical *HTU* values was +15% for hydrogen distillation (fig. 3) and +30% for water distillation (fig. 5). The differences between experimental and theoretical values of heat transfer coefficient are larger (fig. 6). Subcooling reflux at the column top, imperfections of column insulation, and vapor superheating at the column bottom may be the causes of these large differences. This fact requires future improvement of the experiment conditions and corrections of criterion relation coefficients.

Published data regarding other structured packing performances are mostly presented by height of equivalent theoretical plate *HETP*. In order to compare the B7 packing performances with other structured packing performances, we transformed *HTU* in *HETP*, starting from the following equation:

$$Z \quad HTU \quad NTU \quad HETP \quad NTP \qquad (13)$$

where *NTP* means number of transfer plate.

NTP/*NTU* ratio may be calculated using relation [2]:

$$\frac{NTP}{NTU} \quad \frac{1 \quad \frac{j}{a}}{2.3 \lg \quad \frac{a}{j}} \qquad (14)$$

where *j* is the slope of the equilibrium curve and *a* the slope of the operation line.

Figure 7 shows *NTU*/*NTP* ratio variation with *j*/*a* ratio.

At water distillation the curve is almost horizontal, consequently $HETP \cong HTU$. For hydrogen distillation the curve is far from the horizontal line. At low concentrations (<0.1%) $NTP/NTU \cong 1.2$. In both cases *HETP* values lie in range of 0.1-0.24 m. Structured packing manufactured by

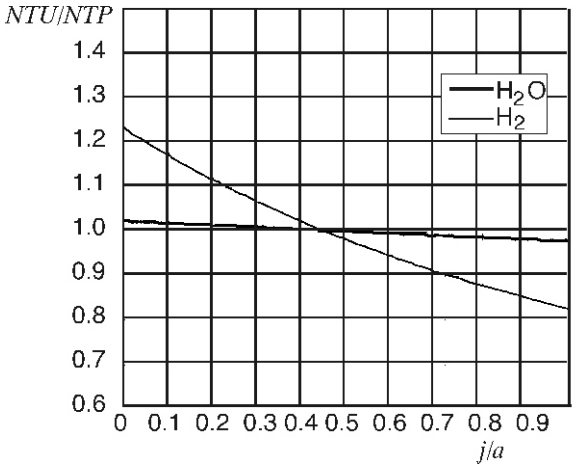

Figure 7. *NTU*/*NTP versus j*/*a*

firms Sulzer and Koch-Glitsch have the same performance (0.15-0.25 m) [7, 8]. Heat transfer coefficient values lie in range of 3-8 W/m^2°C, having the same order of magnitude as the values determined at structured catalyst packing [9].

CONCLUSIONS

The paper presents theoretical and experimental data concerning mass and heat transfer on B7 structured packing, used for deuterium separation in a distillation column. From the results it may be concluded that:

The range of the experimental *HTU* values is 0.1-0.2 m, for both distillation processes, hydrogen and water.

The mathematical model proposed for *HTU* prediction was verified by comparing theoretical and experimental data. The differences between these two kinds of values lie in the range of +15% for hydrogen distillation and in the range of +30% for water distillation.

The differences between the experimental and theoretical values of the heat transfer coefficient are larger. Subcooling reflux at the column top, imperfections of the column insulation, and vapor superheating at the column bottom may be the causes of these large differences. This fact requires future improvement of the experiment conditions and corrections of criterion relation coefficients.

The values of mass and heat transfer coefficient, evaluated theoretically and verified experimentally, are comparable with other structured packing (Sulzer, Koch-Glitsch), used in the chemical industry. This fact justifies the use of a multi-tubular wetted-wall model for describing the transfer processes in distillation columns equipped with B7 ordered packing.

The *HTU* values obtained experimentally or theoretically present the designing data for hydrogen and heavy water plants.

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ИСТРАЖИВАЊА О ПРЕНОСУ МАСЕ И ТОПЛОТЕ Б7 УРЕЂЕНОМ ИСПУНОМ

У раду су приказани теоријски и експериментални резултати проучавања преноса масе и топлоте Б7 уређеном испуном за сепарацију деутеријума у дестилационој колони. Математички модел преноса проверен је експерименталним подацима добијеним из два лабораторијска дестилациона постројења за сепарацију деутеријума. Експериментални подаци о ефикасности Б7 испуне за криогену дестилацију водоника на нивоу од око –250 C добијени су из једног постројења, док су из другог коришћени подаци о преносу масе и топлоте истом испуном за сепарацију деутеријума вакуумском дестилацијом воде на нивоу око 60 C. Вредности висине уређаја за пренос и коефицијената преноса масе и топлоте, одређене теоријски и експериментално, упоредиве су са расположивим подацима о сепарационим поступцима у хемијској индустрији. Ово оправдава употребу вишецевног модела за опис преноса масе и топлоте у дестилационој колони опремљеној Б7 уређеном испуном.