Separation of mixtures substances with similar properties in horizontal rotary mass transfer columns

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Abstract. Information on the use of rotary columns in the processes of fine separation, that is, the separation of substances with similar properties, including isotopes, is systematized. The design features and test results of a new generation of horizontal rotor columns with a rotating mass transfer part are considered in most detail. The results of the use of such columns are presented, both in the rectification of a standard mixture of substances with similar properties, and in the separation of isotopes by the chemical exchange method. A comparison of the results obtained with similar data for classical vertical columns is given, and possible options for the spatial arrangement of horizontal rotor devices are presented.

1. Introduction

A feature of the separation processes of substances with similar properties is a small difference between the single stage separation factor and unity. This leads to the need of a large number of theoretical plates (NTP) and to the use of mass transfer columns of great height [1, pp 1-40]. For example, it is known to use packed columns with a height of several tens of meters for the separation of oxygen isotopes [2, 3] and even (100-200) m in the separation of carbon isotopes by low-temperature distillation of carbon monoxide [4-6]. High column are difficult to manufacture. In addition, the more serious problem is to install them strictly vertically. Moreover, high column needs a high-rise production facilities both aboveground (Figure 1) and underground location [4-6]. The use of horizontal mass transfer apparatus can eliminate such disadvantages and reduce the cost of isotope products. Vertical columns with a rotating mass-transfer unit is usually used in laboratory scale. For example, these are the so-called film columns with a rapidly rotating rotor (Figure 2 a) or a
column with a rotor made of conical elements (Figure 2 b, c), which was used in the separation of oxygen isotopes by distillation of water [8, 9, pp 66-67].

![Figure 2. An example of vertical rotor columns: a - film column; b - view of a conical mass transfer element; c - mass transfer column with conical elements.]

The peculiarity of such columns is a high rotation speed - several thousand revolutions per minute. This should provide a very thin film on the surface of such devices. An example of the use of horizontal mass transfer columns with rotating discs located on a central shaft is known (Figure 3) in the industry. Such columns were used in the concentration of the nitrogen-15 isotope by the chemical exchange method between gaseous ammonia and an aqueous solution of ammonium ion [10]. Such columns were used at the first stage of the production cascade in the former of the USSR.

![Figure 3. An example of a horizontal rotor column of industrial type with a mass transfer part in the form of discs on a central shaft: a – disc element; b - view of the mass transfer part of the column [10].]

A characteristic feature of all the listed rotor devices is a small interface area limited by the geometric area of the used mass transfer elements. The study is devoted to the creation and testing of new types of horizontal rotor columns with a highly efficient phase contact surface.

2. Experimental methods

2.1 Two types of horizontal rotor columns

In our laboratory several types of rotary horizontal columns were tested. The report contains a description of two types of such columns and the results of their testing. It should be noted that the inner diameter of the columns was 30 mm, which often corresponds to the columns of the last stages of the cascades for production of stable isotopes by distillation and chemical exchange methods.

In the column of the first type, the rotating mass transfer part is represented by (Figure 4)
perforated plates 1 on the central shaft 2. The space between the plates is filled with a spiral-
prismatic packing 3 (Levin's packing) [11]. This packing provides a high phase contact surface
(two - three thousand square meters per cubic meter [1, p 31]).

Figure 4. Horizontal column of the first type (type I): 1 - perforated plates located at an angle of
120°; 2 - central shaft; 3 - spiral prismatic packing.

The second type of column is without a central shaft (Figure 5). In this case the packing is placed
in perforated cylinders 1 which have special devices 2 for transferring rotation from one cylinder
to another.

Figure 5. Horizontal column of the second type (type II): 1 - perforated cylinder with special
devices for transferring rotation; 2 - schematic of a device for transmitting rotation.

In fact, all cylinders together are a single mass transfer part of the column.

2.2 Test methods

The study was carried out in two ways. At the first stage, two types (type I and type II) were
investigated in distillation mode using a reference mixture in order to investigate the basic laws
of operation of such columns. A reference mixture is benzene - 1,2-dichloroethane [12, pp 135-
160]. At the same time, which standard mixture is selected based on the expected separation
capacity of the columns.

When testing the columns, the speed of rotation of the mass-transfer part, the angle of inclination
of the column, and the flow of the mixture were changed. At the same time, during the tests, the
change of the pressure drop of the column was studied depending on the rotation speed, the
separating ability of the column, and the mass transfer indicators.

3. Results and discussion

3.1 Distillation mode

The main test results of two types of columns under the reference mixture distillation mode are
shown in Figure 6. The external view of the installation for testing a horizontal rotor column of
type I in the mode of distillation of a reference mixture is shown in Figure 7.

Figure 6. Results of tests of the horizontal rotor columns of the first and second type with the spiral-prismatic packing (the element size 2.5x2.5x0.2 mm) in the mode of distillation of a reference mixture benzene – 1,2-dichloroethane under various conditions (ω - rotation speed of the mass transfer part of the column; \( L_{sp} \) – specific flow of reference mixture; \( γ \) - angle of inclination of the mass transfer part; \( S \) – separation factor):

a - pressure drop of the column without rotation and after the start of rotation of the mass transfer part;
b - change in the separation factor of the reference mixture components without rotation and after the start of rotation;
c - change of the separation factor of the reference mixture components of the over time;
d - change of HTP depending on the specific flow of the reference mixture;
e - influence on the mass transfer of the speed of rotation of the mass transfer part of type I;
f - influence on the mass transfer of the speed of rotation of the mass transfer part of type II.

According to test results (Figure 6) can clearly see the changes that occur in the column at the beginning of the rotation of the mass exchange unit. Pressure drop of the column (Figure 6 a)
increases suddenly and then gradually approaches the steady state value. It is clearly seen that the rotation of the mass exchange unit leads to an increase of pressure drop.

**Figure 7.** The installation for testing a horizontal rotor column of type I in the reference mixture distillation mode.

The separating ability of the column is expressed by the separation factor, that is, the ratio of the concentrations of the target component at the ends of the column. After the beginning of rotation, in spite of the unstable flows and pressure drop in the column, there is an intensive mass transfer. The separation factor $S$ of reference mixture increases rapidly ($C_P$ - concentration of the target component at the bottom, $C_W$ - concentration at the top of the column) – Figure 6 b). Here you can specify that the separation capacity before the start of rotation is equal to the single stage separation factor (it should be recalled that a single stage separation factor for a standard system benzene - 1,2-dichloroethane is equal to 1.08 - 1.09).

At the same time, it is clearly seen from Figure 6 c that, for example, at a speed of 20 rpm for 16 hours after the start of rotation of the mass transfer part of the column, the separation factor $S$ was close to 11.

Using the obtained values of the separation factor $S$ the height of theoretical plate (HTP) was calculated by the method from stage to stage taking into account the dependence of the single stage separation factor from the reference mixture composition. It is interesting that an increase in the flow rate by almost two times does not change the HTP values within the limits of the measurement accuracy of this value (Figure 6 d). Note that similar results were obtained when changing the column tilt angle in the range from 5 to 11 degrees: $\gamma = (8 \pm 3)^\circ$.

On the contrary, as shown empirically, there is a significant effect of the rotation speed on the HTP. For the mass exchange unit of the first type the speed increase ranging from 20 to 60 revolutions per minute leads to a pronounced minimum of the height of theoretical plate (Figure 6 e). The value of HTP is changed three times and minimum of HTP corresponds to speed equal 45 rpm.

For the second type of mass exchange unit (type II) effect of speed on HTP is different (Figure 6 f). When comparing the test results of two types of columns, it should be noted that:
- first, the value of HTP is much smaller because of the greater surface contact between the phases for type II;
- second, the minimum of HTP corresponds to the speed 40 rpm;
- third, an increase in the rate of the mass transfer part of the column leads to a relatively weak growth of the HTP or its absence within the experimental error (all other things being equal).

In general, you must specify that the horizontal rotational column with spiral-prismatic packing has a sufficiently high of mass transfer efficiency: in a length of 80 cm is realized about 25 theoretical plates (Table 1).
Table 1. The results of determining the efficiency of mass transfer for a horizontal rotor column type II with a spiral-prismatic packing (length = 80 cm; $\gamma = 8^\circ$)

| $\omega$, rpm | $L_{SP}$, ml/(cm$^2$·min) | $S$ | NTP | HTP, cm |
|---------------|----------------------|-----|-----|--------|
| 20            | 0.97 ± 0.17          | 5.1 | 16.3 ± 1.3 | 4.9 ± 0.4 |
| 30            | 0.94 ± 0.20          | 6.8 | 19.7 ± 1.6 | 4.1 ± 0.3 |
| 40            | 1.04 ± 0.16          | 10.6| 24.4 ± 1.9 | 3.3 ± 0.3 |
| 50            | 1.05 ± 0.14          | 9.9 | 23.5 ± 1.9 | 3.4 ± 0.3 |
| 60            | 0.86 ± 0.17          | 10.1| 21.4 ± 1.7 | 3.7 ± 0.3 |

So, for a specific flow $L_{sp} \approx 1$ ml/(cm$^2$·min), the separation factor was $S = 10.6$, which corresponds to the NTP value 24 ± 2, and HTP equals 3.3 cm.

Comparing above data with similar results of tests of vertical type columns (Table 2) shows that when a certain combination of parameters such as angle and speed, the efficiency of mass transfer of these types of columns is practically identical in the distillation process.

Table 2. Comparative data on the mass transfer efficiency for classical vertical columns and horizontal rotor columns

| Parameter | Vertical column | Horizontal rotation column |
|-----------|----------------|---------------------------|
| Height (length) packed bed, cm | 60 | 80 |
| Diameter (internal), mm | 30 | 30 |
| The size of the packing, mm | 3x3x0.25 | 2.5x2.5x0.2 |
| Specific flow rate, ml/(cm$^2$·min) | 1.20 ± 0.15 | 1.04 ± 0.16 |
| HTP, cm | 2.9 ± 0.2 | 3.3 ± 0.3 |

3.2 Chemical exchange mode

The determination of the basic conditions for the separation of a mixture substances during distillation mode it possible to proceed to the separation of isotopes by the chemical exchange method. To study the system gas - liquid was chosen, which is a boron trifluoride and its complex with anisole [13-15], which is applied in practice to separate of boron isotopes by isotope exchange reaction as a result of which boron-10 is concentrated in the liquid:

$$^{10}\text{BF}_3\text{(gas)} + ^{11}\text{BF}_3\text{C}_6\text{H}_5\text{OCH}_3\text{(aq)} \rightarrow ^{11}\text{BF}_3\text{(gas)} + ^{10}\text{BF}_3\text{C}_6\text{H}_5\text{OCH}_3\text{(aq)}$$  (1)

In this process flow reflux carried out in accordance with the equation of thermal dissociation of complex for boron-10 concentration (2) and the formation of a complex during heat removal at the concentration of boron-11 (3)

$$^{10}\text{BF}_3\text{C}_6\text{H}_5\text{OCH}_3\text{(aq)} \rightarrow ^{10}\text{BF}_3\text{(gas)} + \text{C}_6\text{H}_5\text{OCH}_3\text{(aq)}$$  (2)

$$^{11}\text{BF}_3\text{(gas)} + \text{C}_6\text{H}_5\text{OCH}_3\text{(aq)} \rightarrow ^{11}\text{BF}_3\text{C}_6\text{H}_5\text{OCH}_3\text{(aq)}$$  (3)

The general scheme of the process of separating boron isotopes is shown in Figure 8, where it can be seen that the complexing agent, anisole, can circulate in a closed technological cycle - Figure 8. This general organization of the process does not depends on the type of column.

Chemical exchange tests were carried out for the second type of column. The column was connected to other flow reflux systems (upper system for the formation of complex compounds, bottom system for the thermal dissociation of complex and return of BF$_3$ to the column.

Figure 8. The general scheme of the boron isotopes separation process by the chemical exchange method (anisole process).
A view of an installation for the separation of boron isotopes by the chemical exchange method with a horizontal rotor column is shown in the Figure 9.

Figure 9. The installation for testing a horizontal rotor column of type II in the mode of boron isotope separation by chemical exchange between BF₃ and its complex compound with anisole.

Column for the separation of boron isotopes has worked under isothermal conditions (40 °C), unlike the distillation process (adiabatic conditions). Experiments were carried out at a speed 40 rpm and at γ = 8°. The change in the concentration of boron isotopes over time during testing of a horizontal column in the chemical exchange mode is shown in Figure 10. The results of experiments showed that the relatively short length of the column there is a significant change in the concentration of boron isotopes at the ends of the column (the difference is about 10% at.).

Figure 10. Change of the concentration of boron-10 isotope at the ends of a horizontal rotor column in time.

The change of the separation factor $S$ in another experiment on the separation of boron isotopes is given in Figure 11.
Figure 11. Change of the separation factor $S$ on the separation of boron isotopes by the chemical exchange in anisole system.

The resulting concentration values correspond to the separation factor $S = 1.84$ and $S = 1.73$ for flow rate of $BF_3 G_{SP} = 0.20$ and $0.34$ mol/(cm$^2$ min).

In general, the separation of boron isotopes by chemical exchange between the $BF_3$ and its complex compound with anisole in the horizontal column, sufficiently good experimental results have been obtained (Table 3). So, on the length of the column 80 cm is reached at about 20 - 27 theoretical plates and HTP value at the same time was (3 – 4) cm.

Table 3. The main results of testing a horizontal rotor column of type II at the separation of boron isotopes by the method of chemical exchange in an anisole system

| Specific flow rate $G_{SP}$, mol $BF_3$/(cm$^2$·h) | Separation factor $S$ | NTP | HTP, cm | Mass transfer factor $K_V$, mmol/(cm$^2$·h) |
|-----------------|----------------|------|--------|------------------|
| 0.20            | $1.84 \pm 0.09$ | 22.9 ± 2.3 | 3.5 ± 0.4 | 57 ± 6         |
|                 | $2.06 \pm 0.14$ | 27.1 ± 2.7 | 3.0 ± 0.3 | 67 ± 7         |
| 0.34            | $1.73 \pm 0.09$ | 20.6 ± 2.1 | 3.9 ± 0.4 | 87 ± 9         |
|                 | $1.80 \pm 0.13$ | 22.1 ± 2.2 | 3.6 ± 0.4 | 94 ± 9         |

Based on the presented results, we can say that the data obtained close to the results of tests of the column in the distillation mode. It can also be said that horizontal rotor columns are effective for separation of boron isotopes by chemical exchange which has already been noted more than once [16, 17].

4. Conclusions

The test results suggest the use of horizontal rotational columns in the industrial separation of boron isotopes, for example. In this case, placement of the columns may be different.

In one case, the columns will be placed so as to arrange for the liquid flow without any additional devices.

In another case, more closely spaced columns require the use of additional pumps for fluid flow. Calculations show that despite the additional cost of electricity for the rotation of the mass exchange unit of columns, the use of such separation devices allows to reduce the cost of obtaining of isotope products about to 1.5 - 2 times. This is due to a significant decrease in capital investment in the creation of high-rise buildings and a reduction in the cost of their operation.

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