POSSIBILITIES FOR REORIENTATION THE ACTIVITY OF HEAVY WATER PLANTS

F. POP, C. CROITORU, GH. TITESCU, I. STEFANESCU,
I. HODOR, S. CUNA**
National R&D Institute for Cryogenics and Isotopes Technologies
ICSI Ramnicu Valcea, Romania, pop.floarea@icsi.ro
National R&D Institute of Isotopic and Molecular Technologies
Cluj – Napoca, Romania, hodor@L40.itim-cj.ro

ABSTRACT

In Romania heavy water is produced by H_2O–H_2S chemical exchange (GS process) and by water distillation, simultaneously working two lines. The distillation plants have high separation capacity, a distillation line being able to concentrate water from two GS lines. The paper presents data regarding possibilities to use one distillation line for oxygen 18 production, as pre-concentrates or finite products. Using a simulation program it was calculated oxygen 18 concentration in heavy water produced, maximum ^{18}O concentration of pre-concentrate obtained on distillation line and the separation cascade dimensions for obtain 95% ^{18}O, with first and second stage having same dimensions like a distillation plant from Romanian heavy water factory. Oxigen-18 separation factor is much lower than deuterium separation factor. For this reason, oxygen-18 is a very expensive product.

Key words: oxygen-18, heavy water, distillation,

1. Introduction

Oxygen-18 is used as tracer in chemistry, in biomedical applications, for environmental and hydrology studies. A very important utilization of oxygen-18 is in medicine, in positron emission tomography (PET) as precursor for ^{18}F production. This utilization has increasing the worldwide demand for ^{18}O as water. Oxygen-18 is market as water at low concentration (10% atom percent) for tracer and at high concentration ($\geq$95% atom percent) for PET. In the last time, the oxigen-18 producers were made public their intention to buy oxygen-18 pre-concentrates.

Oxygen-18 separation is possible by distillation (water, oxygen, carbon monoxide or nitrogen oxide), electrolysis of water, thermal diffusion (oxygen, carbon monoxide), chemical exchange reactions and membrane distillation. In the world were applied water distillation (Rehovoth – Israel), cryogenic distillation (Isotec – USA) and thermal diffusion (Isotec – USA). Water distillation is used for industrial scale. If there is a source of water that has the ^{18}O concentration greater then in natural water, the production cost is lower. This source is the heavy water. Actual worldwide tendency is to turn to good account all existent availability. In Romania, such availability there is at Drobeta Turnu Severin, on heavy water factory and at Ramnicu Valcea, on former heavy water pilot plant.

The paper presents data regarding possibility of the reorientation the heavy water plants activity to oxygen-18 production, in pre-concentrate form or in products form (with 10% and $\geq$95%). In the last case are necessary to make investment expenditure for increase the number of theoretical plates, respectively to increase the height of packing. Enlargement the number of theoretical plants is imposed by the low value of oxygen 18 separation factor in comparison with deuterium separation factor.

2. Oxygen 18 concentration on industrial water isotopic distillation plant

In Romania, at Drobeta Turnu Severin (ROMAG PROD), heavy water is produced by dual temperatures chemical exchange between water-hydrogen sulphide (GS process) and by water distillation. The factory, designed for 360 t/year, has four GS lines. Up to the present, two lines function simultaneously. Fig. 1 shows the actual heavy water production line from Drobeta Turnu Severin [1].

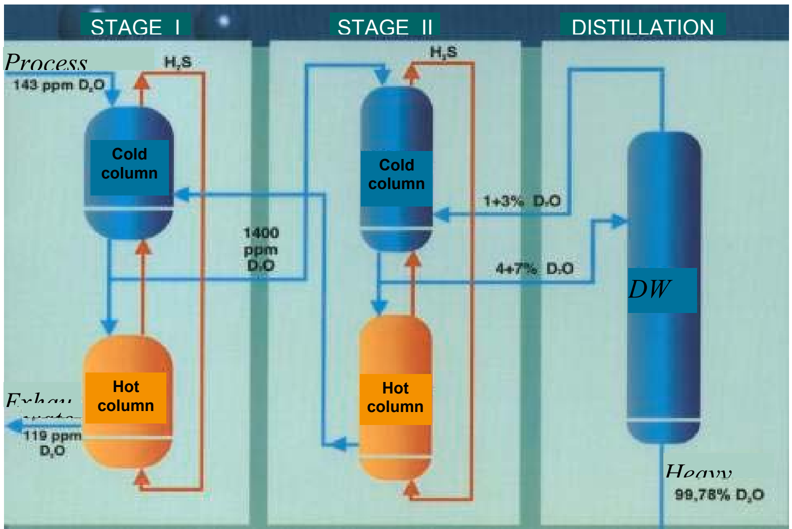

Fig. 1. ***Industrial heavy water production line***

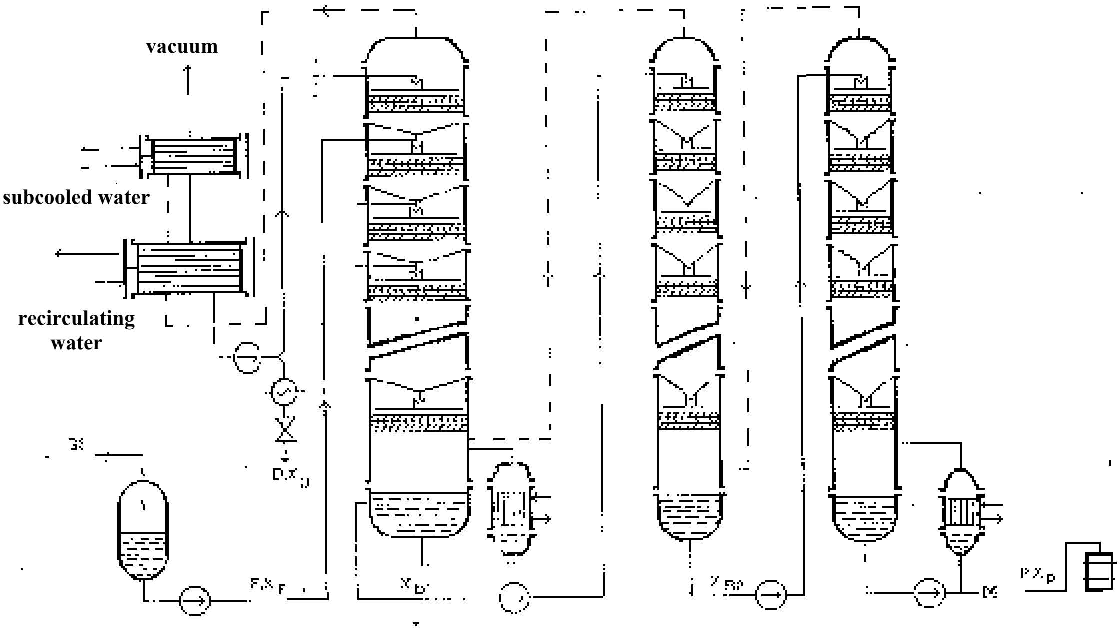

Fig. 2. *Industrial heavy water distillation plant*

The distillation plant, presented in fig. 2, is a separation cascade with two stages [2].

The plant has high separation capacity, a distillation line being able to concentrate water from two GS lines. In this case, one distillation line can be used for oxygen-18 production.
During the heavy water final concentration, by distillation, simultaneously takes place oxygen-18 concentration. Using mathematical simulation program, it was calculated the oxygen-18 concentration in heavy water produced by ROMAG-PROD. Because oxygen-18 concentrates not by H_2O-H_2S chemical exchange, the concentration of distillation feed remains at natural value, 2000 ppm ^{18}O. We used the following work parameters to simulate the distillation plant functioning:

- vapour loading factor (mean value):

stage 1 --> $1.4\ \frac{m}{s}\sqrt{\frac{kg}{m^3}}$ stage 2 --> $1.2\ \frac{m}{s}\sqrt{\frac{kg}{m^3}}$

- temperature (mean value):

stage 1 --> 57.3 ^{O}C stage 2 --> 69.5 ^{O}C

In this conditions oxygen-18 separation factors are: stage 1 – 1.0064, stage 2 – 1.0057. Simulation results show that the heavy water produced by ROMAG PROD contains 0.53% ^{18}O. This value is very closed of the measured concentration, 0.51%. National R&D Institute of Isotopic and Molecular Technologies Cluj – Napoca, Romania Oxygen-18 made mass spectrometry analysis of ^{18}O.
Considering one distillation line being available, it was simulated, on this plant the oxygen-18 pre-concentrates obtaining. Tab. 1 presents the product flow rate (P) as a function of product concentration (x_P) and feeding position. They show that, feeding this plant with heavy water (99.8% D/D+H) of 0.53 % ^{18}O, the maximum concentration obtained is 6%.

Tab. 1 *Flow rates of ^{18}O pre-concentrates / distillation line*

Yp	Feed position								
	1			2			3		
%	kmol/h	g/h	t/an	kmol/h	g/h	t/an	kmol/h	g/h	t/an
2	0.076368	1530	12.24	0.077202	1547	12.38	0.077912	1561	12.49
3	0.038448	771	6.17	0.038823	779	6.23	0.039109	785	6.28
4	0.020218	406	3.25	0.020305	408	3.26	0.020420	410	3.28
5	0.009428	190	1.52	0.009418	189	1.51	0.009347	188	1.50
6	0.002241	45	0.36	0.002107	42	0.34	0.001967	40	0.32

For small product flow rates, deuterium concentration in the waste changes very little, comparatively with the feeding. Table 2 presents these values. Practically all values from table are equal with 99.8%. Increasing de heavy water concentration from 99.80 to 99.84% D/D+H it can be realized the conditions imposed for concentration at first stage-top (Xw = 99.8%). This fact involve the heavy water production decreasing with 0.4%.

Tab. 2 *Waste concentration*

Yp	Feed position		
	1	2	3
%	kmol/h		
2	99.764	99.764	99.763
3	99.783	99.783	99.783
4	99.792	99.792	99.792
5	99.796	99.796	99.796
6	99.799	99.799	99.799

3. Cascade for production oxygen-18 of 95%

Because the ^{18}O separation factor is very low, comparatively with deuterium separation $\left(\alpha_{^{18}O} = 1.006, \quad \alpha_D = 1.05\right)$, it is necessary additional distillation columns to produce oxygen-18 of 95%. Calculus for oxygen-18 separation cascade may be made if between stages are known the

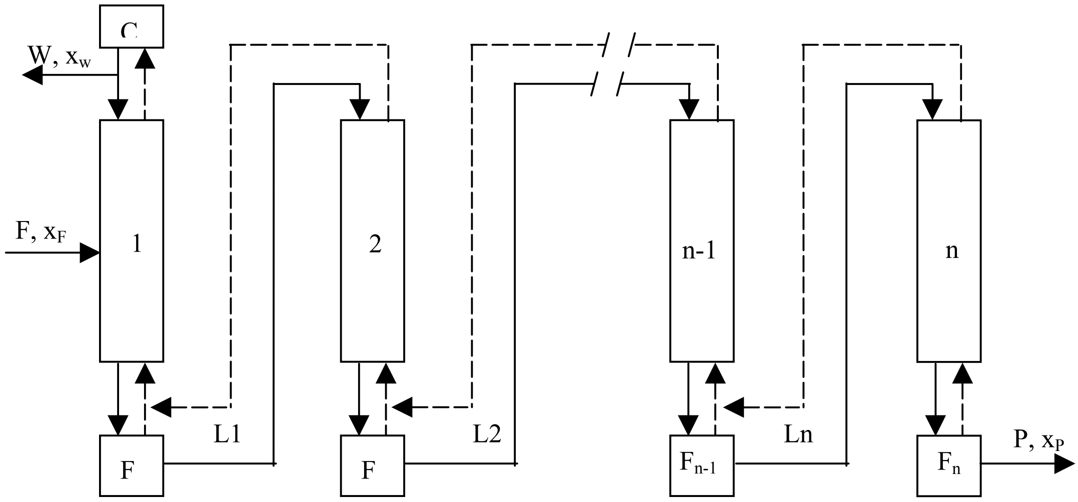

Fig. 3. *Schema of a cascade for isotopic separation*

concentrations or the flow rates. Figure 3 presents separation cascade schema.

Our calculus program, made for this study, imposes the flow rates by the following relation [3]:

$$\frac{L_i}{L_{i+1}} = c \cdot i^k \tag{3}$$

where: L = liquid flow rate, c, k = constant, i = stage number; i = 1 ... n

The cascade that assures at a mean temperature of 50^{O}C oxygen-18 concentration to 95%, using heavy water of 0.53% ^{18}O has six stages. First and second stages of cascade have the same high and diameter with the columns of ROMAG-PROD.

4. Conclusions

Simulation results and mass spectrometry analysis show that on distillation plant from Drobeta Turnu Severin ^{18}O concentration modifies from natural value 0.2% to 0.5%. Calculi also show that the maximum ^{18}O concentration that can be obtained on available distillation line is 6%. Oxygen-18 of 95% can be obtained on a separation cascade with six stages. To produce it, investments are necessary to carry out the four last stage of cascade. ICSI Ramnicu Valcea can take part in this investement with its available isotopic water distillation columns. Taking into consideration that oxygen-18 of 95% is very expensive, 180 $/g (as water), the capitalization of our experience and availabilities is a verry good developement option.

5. References

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