



Validation of the design of small diameter pulsed columns for the process line DRA Tests reliability compared with the industrial scale

J. LEYBROS

DCC/DRRV/SEMP/LGC - CEA, Valrho Marcoule

INTRODUCTION

As part of the SPIN program related to the management of nuclear wastes, studies has been undertaken to develop partitioning processes like DIAMEX process. The process line CCBP/DRA in ATALANTE facility forms one of the main equipment devoted to these studies. The CCBP/DRA purpose is the experimentation of aqueous reprocessing processes implementing representative equipment of industrial apparatus. To this end the use of pulse columns and mixers-settlers was adopted.

The implemented apparatus must be miniaturized taking into consideration requirements related to:

- the facility (ATALANTE) : limiting amount of nuclear matter, gaseous waste minimization, safety, geometrical configuration of the shielded cells;
- the available amount and the costs of the new extractants;
- chemical engineering limiting factors : control of the apparatus working with conditions allowing the scale-up of the results.

From the experimental background from the CEA/FONTENAY-AUX-ROSES and CEA/MARCOULE laboratories, if some knowledge of 25 mm in diameter pulsed columns control were obtained (scale-up studies), there is a little basic studies about 15 mm in diameter pulsed columns working with the system tri-n-butyl phosphate (TBP)- nitric acid - uranyl nitrate.

The decrease in diameter, required for limiting the amount of nuclear matter to be treated, leads to increase the wall effects. It was advisable to demonstrate if that questioned the possibility of scaling up the achieved results.

EXPERIMENTAL

The two air pulsed columns (DN15 and DN25) used for the experiment, whose characteristics went from previous works, were:

- a 15 mm in diameter by 4.4 m high stacked with truncated discs of 20% free area spaced 20 mm and 8 mm in diameter pulse leg.
- a 25 mm in diameter by 4.5 m high stacked with truncated discs of 26% free area spaced 25 mm and 14 mm in diameter pulse leg.

Each column was run in the organic continuous mode (common in industrial plant), for the whole tests.

The test variables were total superficial throughput (aqueous plus organic superficial flow rates - TST), aqueous-to-organic flow ratio (A/O), direction of mass transfer (dispersed to continuous $d \rightarrow c$ or continuous to dispersed $c \rightarrow d$) and the pulsation intensity (amplitude-frequency product $A_p f$).

Fluids were transferred by gear pumps or by gravity. The flow rates were controlled by weighing with precision scales.

Operation of the system was controlled by a microcomputer.

For the whole test the solvent used was 30 vol.% TBP in hydrogenated tetrapropylene (TPH)

The systems used were 30 vol.% TBP-nitric acid-water and 30 vol.% TBP- nitric acid - uranyl nitrate.

The experiment lasted 6 to 10 hours, depending on total throughput, to reach steady-state condition. Uranium in the aqueous and organic phases was analyzed by mean of ICP, and nitric acid by volumetric titration.

RESULTS AND DISCUSSION

The comparison of the two columns is based on three main criteria, which define the pulsed column characteristics:

- pulsation capability : for an air pulsed column, the reachable amplitude-frequency product ($A_p f$) depends on air feed line geometry and particularly it depend on column packing hydrodynamic characteristics (friction coefficient K_f and kinetic coefficient K_k);
- superficial throughput : It depends on discs geometry (spacing and fractional free area) and physicochemistry properties of the two phases;
- mass transfer efficiency: it depends on two opposed criteria, the interfacial area and the driving force to be maintained as high as possible throughout the column. An increase in interfacial area (by increase in pulsation intensity) lead to an increase in axial dispersion (as a consequence, the concentration gradient is degraded throughout the column).

Pulsation capability : The pulsation capability of DN25 column is slightly better than the DN15 column as shown in figure 1. The achieved values confirm the equivalence of the two columns compared with this criterion.

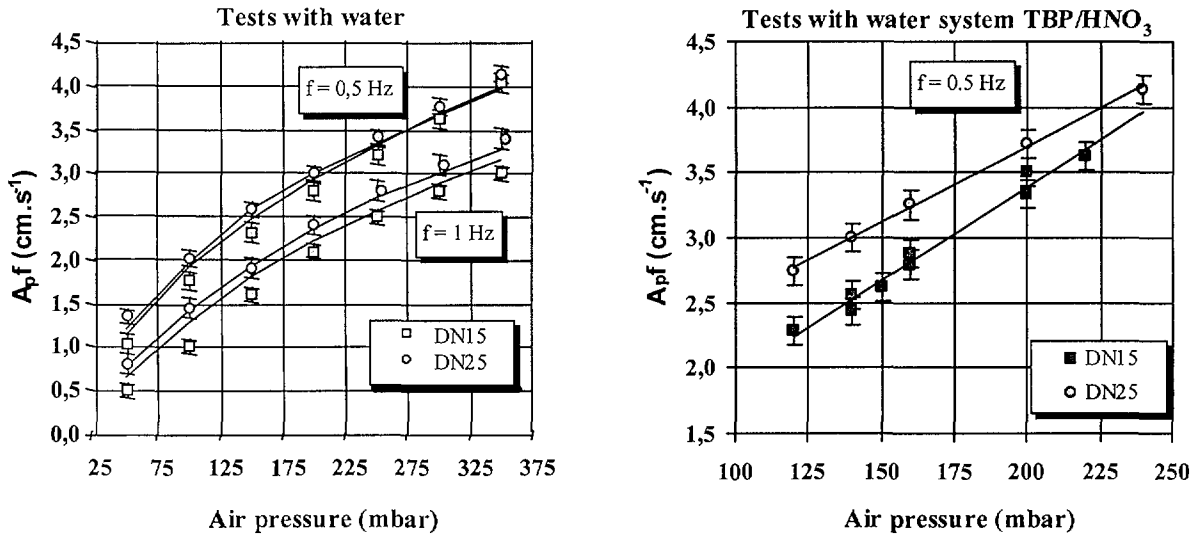


Figure 1 : Pulsation capability

Superficial throughput : The superficial throughput is related to the dispersed phase hold up. The hold up in this study is defined as the average per cent of the total volume of column occupied by the dispersed phase at steady state operation. Aqueous hold up data were obtained at several different continuous-dispersed flow rates and a range of pulsation intensity (1.5 – 4.5 cm.s⁻¹). Representative results of hold up are shown in figure 2, for 30 vol.% TBP equilibrated with nitric acid 1N. These figures demonstrate that the hold up of DN15 column is always higher, for a same pulsation intensity, compared with DN25 column. The difference can be related to the wall effects, which are more intense with DN15 column.

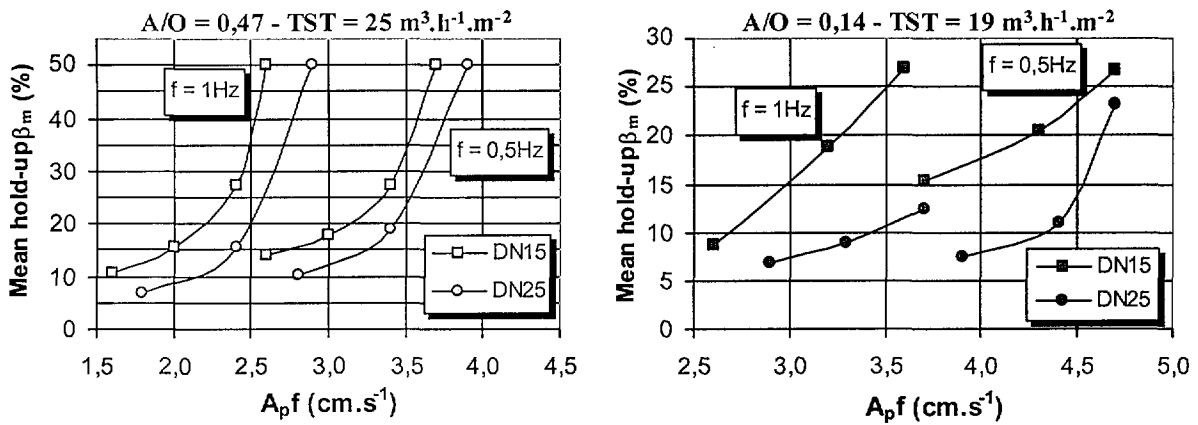


Figure 2 : Effect of pulsation intensity on hold up - system 30% vol.TBP - HNO₃

Mass transfer efficiency : To be compared, all the tests were run with a pulsation intensity such as the mean dispersed phase hold up (β_m) being similar in each column.

A first set of tests was conducted with the system 30 vol.% TBP-nitric acid-water. Extraction tests were conducted with an aqueous feed solution consisting of 3.5 or 5.5 M nitric acid. Stripping tests were conducted with an organic feed solution consisting of 0.65 M nitric acid in 30 vol.% TBP and deionized water as stripping solution. The working conditions are listed in tables I and II. Results were analyzed in term of axial dispersion coefficients (E_c and E_d) using the subroutine PAREX, based on diffusion model and of height equivalent to theoretical stage (HETS) using the McCabe & Thiele method. The calculated values are resumed in figure 3. For the two columns, in similar working conditions, the HETS vary between 0.39 and 0.63 m. The mean value for the DN15 column is about 0.5 m and for the DN25 column the mean value is about 0.54 m.

The mean values for axial dispersion coefficients are the following :

- DN15 column E_c = 6.0 cm².s⁻¹ E_d = 17.0 cm².s⁻¹
- DN25 column E_c = 8.0 cm².s⁻¹ E_d = 20.5 cm².s⁻¹

N°	DN	A/O	TST	$A_p f$	β_m
A1	15	0,20	1,50	3,9	18,1
	25			3,7	14,9
A2	15	0,14	1,90	4,5	21,5
	25			3,6	24,5
A3	15	0,25	2,00	3,5	12,2
	25			3,1	10,9
A4	15	0,25	2,02	3,6	26,2
	25			3,1	19,9
A5	15	0,20	2,02	4,5	14,6
	25			3,5	11,5
A6	15	0,25	2,04	3,0	15,7
	25			3,4	12,0

Table I : Extraction tests working conditions
 – system 30 vol.% TBP-nitric acid-water -
 d→c mass transfer - $f = 0,63$ Hz

N°	DN	A/O	TST	$A_p f$	β_m
S1	15	0,20	1,42	3,0	9,0
	25			3,5	10,8
S2	15	0,30	1,50	2,9	15,7
	25			3,7	11,8
S3	15	0,20	1,96	3,0	11,2
	25			3,2	10,8
S4	15	0,20	1,96	3,0	12,6
	25			3,7	10,8
S5	15	0,25	2,00	3,9	12,0
	25			3,1	10,8
S6	15	0,30	2,20	3,8	17,1
	25			3,1	16,5

Table II : Stripping tests working conditions
 – system 30 vol.% TBP-nitric acid-water -
 c→d mass transfer - $f = 0,63$ Hz

A second set of tests was conducted with the system 30 vol.% TBP- nitric acid - uranyl nitrate. Extraction tests were conducted with an aqueous feed solution consisting of 180 – 195 g.l⁻¹ uranium in 0.7 M (U1 to U3) or 3 M (U4 & U5) nitric acid. The working conditions are listed in table III. Results were analyzed in term of axial dispersion coefficients (E_c and E_d) using the PAREX subroutine. The comparison of the uranium concentration in aqueous raffinate shows that the global efficiency of DN15 column is better than DN25 column. The efficiency strongly depends on TST value. The retained values for axial dispersion coefficients are the following :

- DN15 $E_c = 6.5 \text{ cm}^2.\text{s}^{-1}$ $E_d = 17.0 \text{ cm}^2.\text{s}^{-1}$
- DN25 $E_c = 11.0 \text{ cm}^2.\text{s}^{-1}$ $E_d = 22.5 \text{ cm}^2.\text{s}^{-1}$

These values confirm those previously obtained for the DN15 column. The value of E_c for DN25 column is higher.

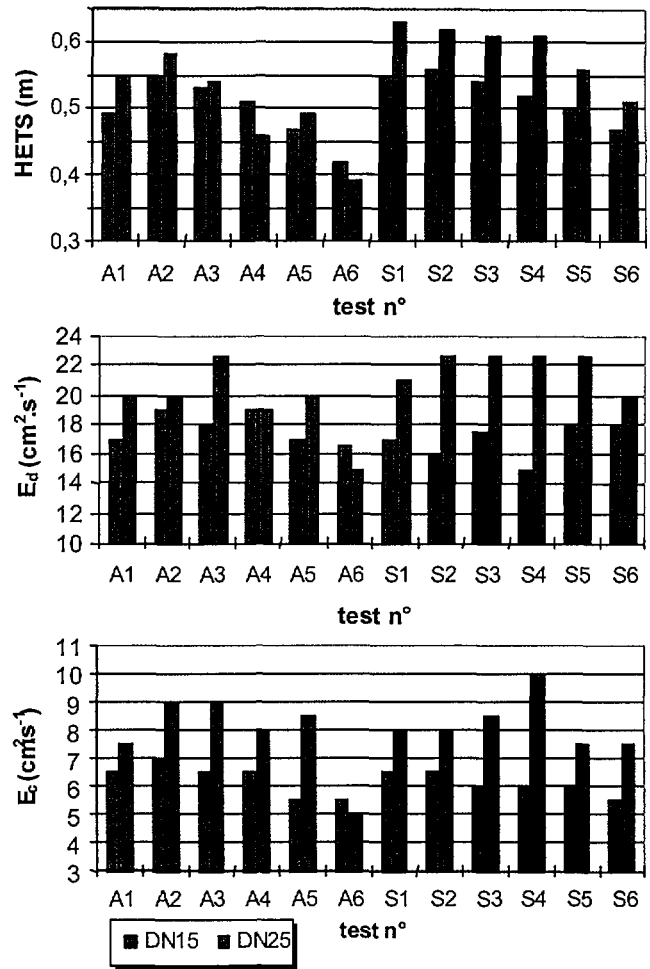


figure 3 : values of HETS, E_c and E_d

N°	DN	TST	$A_p f$	β_m	$[U]_R$	E_d	E_c
U1	15	0,63	3,36	15,1	9,6	17	6,0
	25	0,60	4,13	13,9	210	22,5	11,0
U2	15	1,25	2,79	18,1	0,8	17	7,0
	25	1,21	3,26	14,2	1,9	22,5	10,5
U3	15	2,50	2,27	28,6	1,7	60	6,5
	25	2,45	2,74	31,2	1,0	50	10,5
U4	15	1,01	3,62	26,1	0,03	17	7,0
	25	1,00	4,13	22,5	0,5	22,5	13,5
U5	15	2,52	2,43	32,5	0,04	85	5,5
	25	2,50	3,00	31,9	0,17	90	13,5

Table III : Extraction tests working conditions
 System 30 vol.% TBP- nitric acid - uranyl nitrate
 dAc mass transfer - $f = 0,63$ Hz - $[U]_R$ uranium
 concentration in aqueous raffinate (mg.l⁻¹)

The values of E_c and E_d were used to calculate the concentration profiles of tests n°U1 as show in figure 4. The concentration profiles calculated with the model are in most cases very close to experimental values when using the mean values of axial dispersion coefficients.

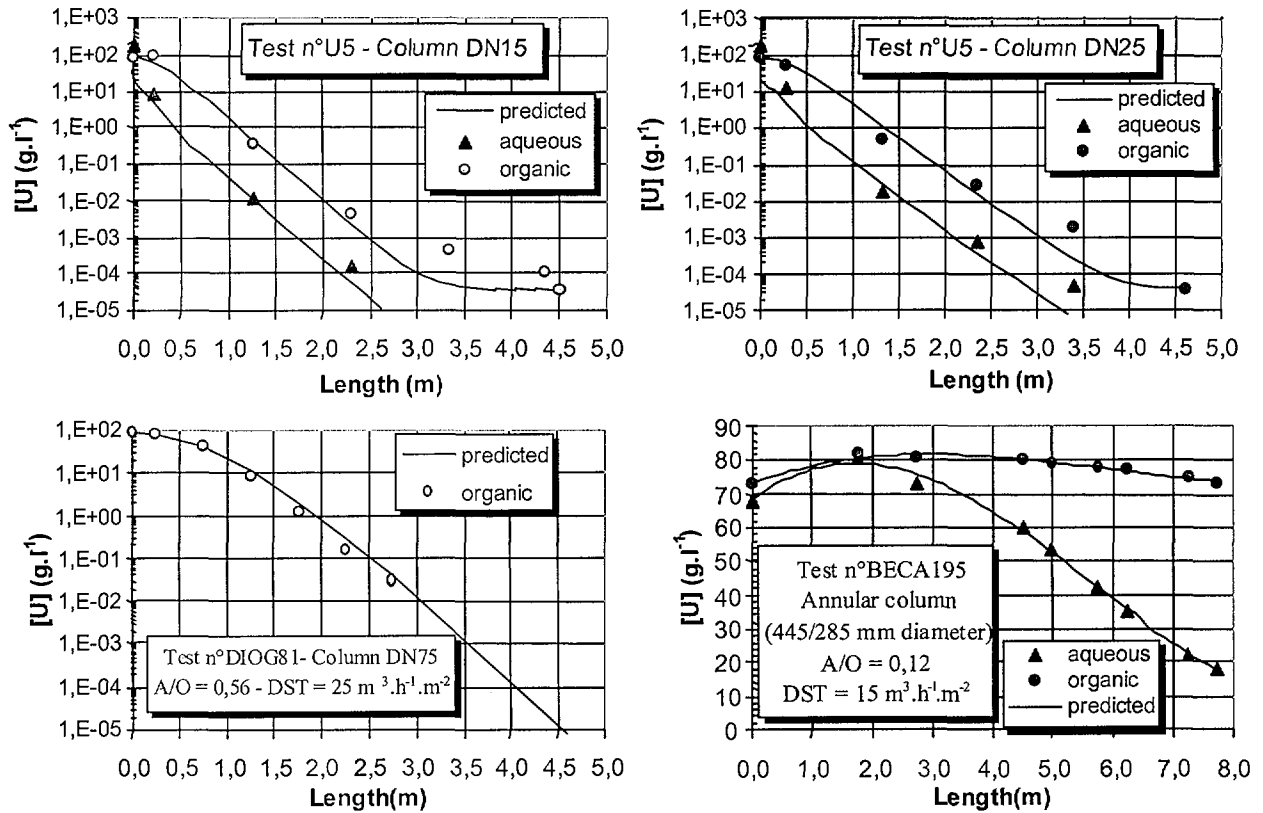


Figure 4 : Extraction profiles for various columns

These profiles can be compared with other experimental and calculated profiles obtained with largest columns. Figure 4 show an organic profile obtained with a 75 mm in diameter column working in extraction conditions (organic continuous mode - $E_c = 12.5 \text{ cm}^2.\text{s}^{-1}$ - $E_d = 34.0 \text{ cm}^2.\text{s}^{-1}$) and with an annular pulsed column, with the same characteristics of the extraction column of the La Hague reprocessing plant, working in stripping conditions (aqueous continuous mode - $E_c = 10.0 \text{ cm}^2.\text{s}^{-1}$ - $E_d = 5.5 \text{ cm}^2.\text{s}^{-1}$). The comparison of experimental and calculated concentration profiles is also satisfactory as demonstrate previously (1).

CONCLUSIONS

It is possible to calculate the concentration profiles of a DN15 column or of an industrial column when the two axial dispersion coefficients are known.

The overall comparison shows that a pulsed column of 15 mm in diameter can be considered as a representative tool of research and development. Particularly, the study demonstrates the possibility of scaling up the results.

REFERENCE

1. P.BARON & J.DUHAMET "Simulation of uranium/plutonium splitting in a pulsed colum in the PUREX process", ISEC'88, Int.Solv.Extr.Conf. Moscow, (IV), 204, (1988)