TESCE, July 2007, pp. 53 -71

Sharp Separation Systems Synthesis: Advances in Combinatorics

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Abstract

The analysis of combinatorics of sequences of sharp separation systems is a well-studied problem in the field of chemical engineering: first Thompson and King (1972) present a closed-form expression for determining the number of possible separation sequences for separating an n-component mixture into pure products by using simple (one input and two outputs) sharp separators. Then, Shoaei and Sommerfeld (1986) show that this determination could be interpreted in term of Catalan numbers; and recently Wahl and Lien (1990) derive this formula form a generating function. This paper presents a closed-form expression for the number of different possible separation sequences when complex (one input and three or more outputs) separators with two or three outputs are used, and finally a generating function of the number of distinct complex separators is given.

Sequences of sharp separators: recursive producers

The design of sharp separation sequences is one of the most investigated problem in the synthesis of chemical units. It consists in generating all different possible separation schemes when an n-component mixture has to be separated into pure products, with the main assumptions:

- only sharp separators are used, i.e. each component of the feed stream exists in only one output stream of the separator;
- the components are ranked in any stream;
- this ranked list of components is invariable;
- mixture or division of intermediate streams is prohibited.

The number of separation sequences may be defined recursively for sequences involving complex sharp separators (Shoaei and Sommerfeld, 1986; Wahl and Lien, 1990; Domenech et al., 1991). For the case of sharp two-output separators (see Figure 1), the number S_n of distinct sequences for an n-component fed-stream may be stated as:

$$\left| \begin{array}{l} S_0 = 0 \\ S_1 = 1 \\ \\ S_n = \sum_{K=1}^{n-1} S_k S_{n-k} \ldots n \geq 2 \end{array} \right. \qquad (1)$$

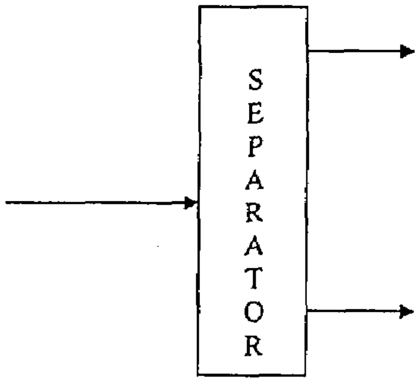

Figure 1: A sharp two-output separator

A three-output separator can be illustrated by a distillation column involving a side stream (see figure 2).

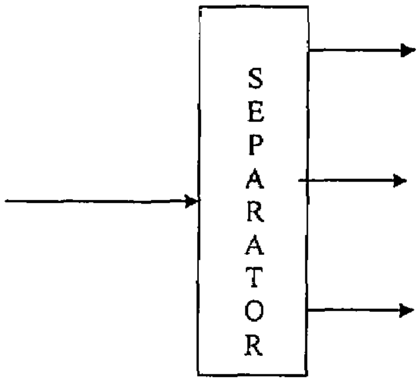

Figure 2: A sharp three-output separator

The earlier works of Petlyuk et al. (1965), Elaahii and Luyben (1983) or Alatiqui and Luyben (1985) show the interest of this type of separator in practice, for small values of n as a matter of fact. The number of possible separation schemes, when each sharp separator is allowed to have either two or three outputs, is then (Wahl and Lien, 1990; Domenech et al., 1991) defined by:

$$\left| \begin{array}{l} S_0 = 1 \\ S_1 = 1 \\ S_n = \sum_{K=1}^{n-1} S_k . \sum_{j=0}^{n-k-1} S_j . S_{n-k-j} \ldots . n \geq 2 \end{array} \right. \qquad (2)$$

In the general case, the minimum number of separators used to separate n components to be achieved is theoretically one and the maximum number is n-1 (in that last case, only simple sharp separators are used). Then, the number of possible separation schemes is formulated by the relation below (Domenech et al., 1991):

$$\begin{cases} S_0 = 1 \\ S_1 = 1 \\ S_n = \sum_{i=1}^{n-1} S_i . S_{n-i} + \sum_{k=1}^{n-2} \sum_{i1} \sum_{i2} \dots \sum_{i_{k+1}} S_{i1} . S_{i2} \dots S_{ik+1} . S_{n-i1-i2} - \dots - ik + 1 \end{cases} \qquad (3)$$

with

$$\begin{aligned} & i1 \in [1, n-1-k] \\ & i2 \in [1, n-1-(k-1)-i1] \\ & \dots\dots\dots\dots \\ & i_{k+1} \in [1, n-1-i1-i2-\dots\dots-ik] \end{aligned}$$

Sequences of sharp separation: closed form formulae

Thompson and King (1972) first presented a closed form expression for Sn for sequences involving only simple sharp separators:

$$S_n = \frac{(2(n-1)!}{n!(n-1)!} \qquad (4)$$

Shoaei and Sommerfeld (1986) pointed out that this determination is an application of Catalan numbers.

When two-or-three-output separators are admitted a closed form equation can be derived (Floquet et al, 1991):

$$S_n = \sum_{i=0}^{E\left(\frac{n-1}{2}\right)} \frac{(2n-2-i)!}{i!n!(n-2i-1)!} \qquad (5)$$

where the function E(x) represents the bracket function. It can be noted that:

$$S_n = \frac{(2(n-1)!}{n!(n-1)!} + \sum_{i=1}^{E\left(\frac{n-1}{2}\right)} \frac{(2n-2-i)!}{i!n!(n-2i-1)!} \qquad (6)$$

The first term is equivalent to the number of sequences found by Thompson and King formula and the second term corresponds to the number of sequences where i three-output separators can appear. The function E expresses that for the separation of an n-component mixture, the maximum number of three-output column is equal to $E\left(\frac{n-1}{2}\right)$.

The formulation of a closed form expression, when the number of outputs for each separator is not specified, is a rather difficult task (Floquet et al.,1991). For an n-component mixture, it leads to:

$$S_n = \sum_{mk\in In} \frac{\binom{n+m-1}{m-1}}{m} \prod_{mj\in In} \binom{m-m_{n-2}-....-m_{j+1}}{m_j} \qquad (7)$$

where

$$In = \{m_{n-2}, m_{n-3}, ..., m_1, m_0 \,/\, m_0 + 2m1 + ... + (i+1)mi + ... + (n-1)m_{n-2} = n-1\}$$

$$m = \sum_{mk\in In} m_k$$

Then :

$$S_n = \sum_{mk\in In} \frac{(n+m-1)!}{n! \prod mk!} \qquad (8)$$

with the same definition of In and m.

The main feature of these expressions is the definition of the set I_n of admissible structures of sharp separation for n components. A structure is a set of sequences involving the same number of same type separators. For example, the two following sequences belong to the same structure (made up of two simple separators and a three-output column):

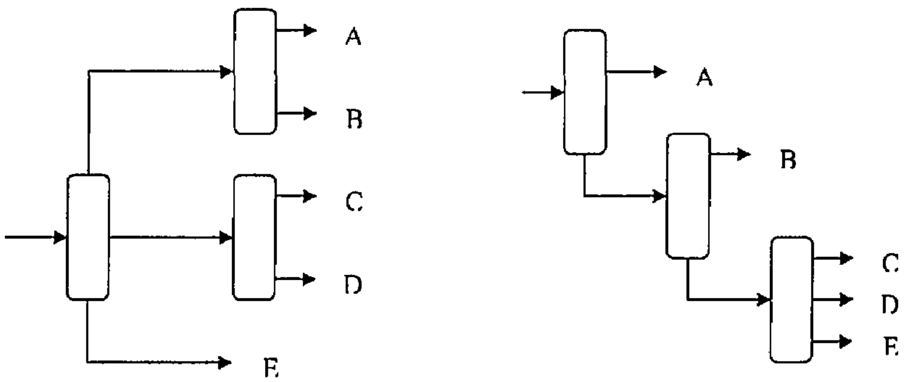

Figure 3a: Two sequences of a same structure

and the following one belong to a different structure:

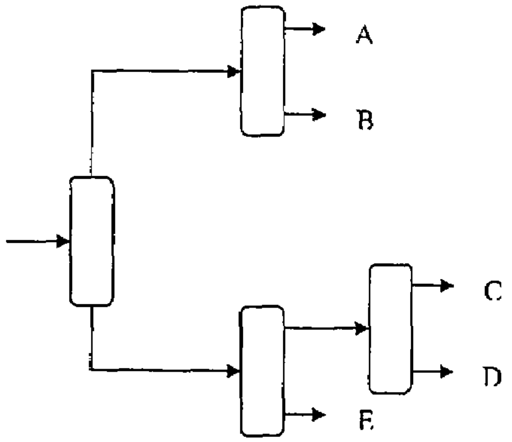

Figure 3b: Two sequences of a different structure

The definition of I_n involves an integer relation between the number mk of possible separators with (k+2) outputs. When the number of outputs is limited to two (simple separators) or three, then the definition of I_n is a trivial task. It becomes:

$m_0 = n-1$ and $m = m_0 = n-1$ for simple separators (the number of simple sharp separators for separating an n-component mixture is n-1) and relation (8) is then equivalent to (4);

$m_0 + 2m_1 = n-1$ and $m = m_0 + m_1$ for two-or-three output separators. The substitution of m_0 by $n-1-2m_1$ in (8) leads to (5).

For complex separators (more than three outputs), the use of equation (8) needs the resolution in the space of integer numbers of:

$$m_0 + 2m_1 + \ldots + (n-1)m_{n-2} = n-1 \qquad (9)$$

The enumeration of all integer solutions of such an equation for an important value of n is not an easy work. However, we can enumerate the total number n_s of elements of In corresponding to an equation of this type, i.e. the number of distinct structures of separation schemes. The main enumeration steps are the following:

1st step: calculate the number of integer solutions of equation
$m_0 = n-1$

n	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
n_s	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1

Table1: number of integer solutions of $m_0 = n-1$.

2nd step: calculate the number of integer solutions of equation
$m_0 + 2m1 = n-1$, i.e. repeat the 1st step with right shifts of two positions.

n	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	....
$m_1=0$	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
$m_1=1$			1	1	1	1	1	1	1	1	1	1	1	1	1	1
$m_1=2$					1	1	1	1	1	1	1	1	1	1	1	1
$m_1=3$							1	1	1	1	1	1	1	1	1	1
$m_1=4$									1	1	1	1	1	1	1	1
$m_1=5$											1	1	1	1	1	1
$m_1=6$													1	1	1	1
$m_1=7$															1	1
n_S	1	1	2	2	3	3	4	4	5	5	6	6	7	7	8	8

Table2: number of integer solutions of $m_0 + 2m_1 = n-1$.

<u>3rd step:</u> calculate the number of integer solutions of equation
$m_0 + 2m1 + 3m_2 = n-1$, i.e. repeat the 2nd step with right shifts of three positions.

n	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	....
$m_1=0$	1	1	2	2	3	3	4	4	5	5	6	6	7	7	8	8
$m_1=1$				1	1	2	2	3	3	4	4	5	5	6	6	7
$m_1=2$							1	1	2	2	3	3	4	4	5	5
$m_1=3$										1	1	2	2	3	3	4
$m_1=4$													1	1	2	2
$m_1=5$																1
ns	1	1	2	3	4	5	7	8	10	12	14	16	19	21	24	27

Table3: number of integer solutions of $m_0 + 2m1 + 3m_2 = n-1$

<u>4th step and others</u>: the calculation of the number of integer solutions of equation
$m_0 + 2m1 + 3m_2 + 3m_4 = n-1$ leads to the following table. The procedure is repeated until the total desirable calculation.

n	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	....
$m_3=0$	1	1	2	3	4	5	7	8	10	12	14	16	19	21	24	27
$m_3=1$					1	1	2	3	4	5	7	8	10	12	14	16
$m_3=2$									1	1	2	3	4	5	7	8
$m_3=3$													1	1	2	3
ns	1	1	2	3	5	6	9	11	15	18	23	27	34	39	47	54

Table4: number of integer solutions of $m_0 + 2m1 + 3m_2 + 3m_4 = n-1$

The number n_S can easily be found with this above procedure, for example by using a spreadsheet. It can also be directly computed with a recurring kth-order equation, where k is the index of the last m_i in the left hand side term of equation (9) and the periodicity is the lowest common multiple of 1,2,3,....,k. for example, the number n_S of solutions of $m_0 + 2m_1 = n-1$ is:

$n_S = A(n-1)-B$; where A=1/2 an B = 1 and 1/2 alternatively

And the number of solutions of $m_0 + 2m1 + 3m_2 = n-1$ is :

$n_S = A(n-1)^2 + B(n-1) + C$; where A = 1/12, B=1/2 and C=1, 5/12,2/3,3/4,2/3,5/12 and 1 successively.

The detailed procedure for determining the values of A,B,C,... is exposed in Appendix I. the values of ns as a function n are given in the following table:

n	n_S
2	1
3	2
4	3
5	5
6	7
7	11
8	15
9	22
10	30

Table5: number of structures of sharp separators.

Finally, the number of sequences of sharp separators is summarized in table6:

Number of components 2	Number of sequences of simple separators (one input-two outputs)	Number of sequences of two or three output separators	Number of sequences of complex (from two to n outputs) separators
2	1	1	1
3	2	3	3
4	5	10	11
5	14	38	45
6	42	154	197
7	132	654	903
8	429	2871	4279
9	1430	12925	20793
10	4862	59345	103049

Table6: number of sequences of sharp separators

sequences of sharp separators: generating function use

The use of generating functions is an elegant way to derive Thompson and King formula (1) (or (4)) and it is very often described in combinatorics textbooks. It consists in determining the infinite power series expansion of a given function that supplies the coefficients S_n of Catalan numbers. The generating function for equation (1) (or (4)) may be expressed as:

$$g(x) = S_0 + S_1x + S_2x^2 + ...+S_nx^n + ... \qquad (10)$$

where $S_0 = 0$; $S_1 = 1$

and S_n ($n \geq 2$) given by eq. (1) or (4)

then, (Wahl and Lien, 1990):

$$g(x) = \frac{1-\sqrt{1-4x}}{2} \qquad x \le 1/4 \qquad (11)$$

is solution of eq. (10).

For the case of two or three separation sequences, the generating function g(x) is:

$$g(x) = S_0 + S_1x + S_2x^2 + \ldots\ldots + S_nx^n + \ldots (12)$$
$$= 1+x+\ldots\ldots+S_nx^n+\ldots$$

where $S_0 = 0$; $S_1 = 1$

and S_n $(n \ge 2)$ done by eq. (2) or (5)

In order to initialize the recursive formula (10) and (12), the values of S0 and S1, without physical sense, were chosen.

Then, (Wahl and Lien, 1990) g(x) is proved to be one of the real solution of the third degree equation:

$$g^3(x)-2g^2(x)+x+1=0 \qquad (13)$$

The first step of the solution of this classical equation is the elimination of the term $-2g^2(x)$:

$$g(x) = f(x) + 2/3 \qquad (14)$$

So, the equation (13) is then equivalent to:

$$f^3(x) - \frac{4}{3}f(x) + (x + \frac{11}{27}) = 0 \qquad (15)$$

Solving this equation (15) for the case $x \in]-1,5/27[$ (functions f and g are defined for $x = 0$) leads to three real roots:

$$\left|\begin{aligned} f1(x) &= \frac{4}{3}\cos\varphi \\ f2(x) &= \frac{4}{3}\cos(\varphi + \frac{2\pi}{3}) \\ f3(x) &= \frac{4}{3}\cos(\varphi - \frac{2\pi}{3}) \\ with\,\varphi &= \frac{1}{3}\arccos(\frac{-(27x+11)}{16}) \quad 0<\varphi<\pi \end{aligned}\right. \qquad (16)$$

From equations (12) and (14) it follows that f(0) = 1/3 and \f(df;dx)(0) = 1, and therefore solutions $f_2(x)$ and $f_3(x)$ can be eliminated. Thus, the generating function g(x) is:

$$\left|\begin{aligned} g(x) &= \frac{4}{3}\cos\varphi + \frac{2}{3} \\ with\,\varphi &= \frac{1}{3}\arccos(\frac{-(27x+11)}{16} \quad 0<\varphi<\pi \\ &and\, x \in]-1,5/27[\end{aligned}\right. \qquad (17)$$

The equivalence of equations (17) and (5) appears to be very hard to strictly derive, however we can compute the first elements of the power series expansion for checking that equation (5) can be generated by the function g(x) given in (17).
From Mac Laurin's series expansion:

$$g(x)\ g(0) + \frac{g'(0)}{1!}x + \frac{g''(0)}{2!}x^2 + \ldots + \frac{g^{(n)}(0)}{n!}x^n + \ldots \quad (18)$$

where $g^{(n)}(0) = \frac{d^n g}{dx^n}(0)$

It comes $g(0) = 1$ from (13) $\Rightarrow S_0 = 1$ in (12)

$$g'(x) = \frac{1}{g(x)(4-3g(x))} \text{ from (12)} \Rightarrow g'(0) = 1 \Rightarrow S_1 = 1 \text{ in (12)}$$

$$g''(x) = \frac{2(3g(x)-2)}{(g(x)(4-3g(x)))^3} \Rightarrow g''(0) = 2 \Rightarrow S_2 = 1$$

$$g'''(x) = \frac{6(15g^2(x)-20g(x)+8)}{(g(x)(4-3g(x)))^5} \Rightarrow g'''(0) = 18 \Rightarrow S_3 = 3$$

......

$$g^{(n)}(x) = \frac{n!}{(g(x)(4-3g(x)))^{2n-1}} D_n(g(x)) \Rightarrow S_n = D_n(g(0)) = D_n(1)$$

where $D_n(g(x))$ is a (n-1) degree polynomial expression of g(x) with

$$\begin{cases} D_1(g(x)) = 1 \\ D_{n+1}(g(x)) = \dfrac{D'_n(g(x))(g(x)(4-3g(x))^2 + 2(2n-1)D_n(g(x))(3g(x)-2))}{n+1} \\ \textit{for} \quad n \geq 2 \quad \textit{the proof is given in Appendex B} \end{cases} \quad (19)$$

These relations give another recursive expression (distinct from relation (2)) for determining the number S_n of sequences with two-or-three-output column for separating an n-component mixture. Table 7 shows the numerical values obtained from relation (19); it can be observed that the values of S_n given in table 7 are the same that those reported in the third column of Table 6. So, from this indirect derivation, the equivalence of equations (17) and (5) can be admitted.

Number of components n	Constant coefficient in $D_n(g(x))$	Coefficient of x in $D_n(g(x))$	Coefficient of x^2 in $D_n(g(x))$	Coefficient of x^3 in $D_n(g(x))$	Coefficient of x^4 in $D_n(g(x))$	$S_n = D_n(g(0)) = D_n(1)$
1	1					1
2	-2	3				1
3	8	-20	15			3
4	-40	140	-180	90		10
5	224	-1008	1812	-1584	584	38

Table7: number of sequences of separation given by relation (19)

Number of distinct sharp separators: generating function use

From Table 6, it can be seen for example that the four-component problem has five different sequences of simple separators. Each sequence involves three separators, giving 15 separators all in all. In fact, there are only 10 distinct simple separators (see Table 8). In the same manner, there are 10 sequences of two-or-three different separators for the four-components problem, but only 15 distinct separators. The number of distinct separators, as noted by Wahl and Lien (1990) $TS(n)_r$ for an n-component mixture to be separated with r-output separators, is important in practice because it gives a lower bound on the total computational effort.

n	Distinct separators	number
n=2	A/B	
n=3	A/BC;AB/C	
	A/B;B/C	
	A/B/C	
n=4	A/BCD;AB/CD;ABC/D	
	A/BC;AB/C;B/CD;BC/D	
	A/B;B/C;C/D	
	A/B/CD;A/BC/D;AB/C/D	
	A/B/C;B/C/D	

Table8: Distinct separators for an- n-component mixture (two-or-three output separators)

The values of $TS(n)_r$ and TS(n) (TS(n)represents the total number of distinct separators, having from two to n output, for separating an n-component mixture) given by Wahl and Lien (1990) are the following:

$$\left|\begin{array}{l} TS(n)_2 = \binom{n+1}{3} = \dfrac{n^3 - n}{6} \\ TS(n)_3 = \binom{n+1}{4} = \dfrac{n^4 - 2n^3 - n^2 + 2n}{24} \\ \dots\dots\dots\dots \\ TS(n)_r = \binom{n+1}{r+1} \end{array}\right. \qquad (20)$$

and $TS(n) = \sum_{k=3}^{n-1}\binom{n+1}{k}$ (21)

From table 8, it can be proved recursively (see Appendix 3) that the values of $TS(n)_r$ and TS(n) can also be expressed by:

$$\begin{cases} TS(n)_2 = \sum_{i=1}^{n-1} i(n-i) & n \geq 2 \\ TS(n)_3 = \sum_{i=1}^{n-2} \frac{i(n-i)(n-i-1)}{2} & n \geq 3 \\ \dots\dots\dots\dots\dots\dots & \\ TS(n)_r = \sum_{i=1}^{n-r+1} \frac{i(n-i)\dots(n-i-r+2)}{(r-1)!} & n \geq r \end{cases} \quad (22)$$

Then : $TS(n)_r = \sum_{i=1}^{n-(r-1)} \frac{i(n-i)!}{(n-i-(r-1))!(r-1)!}$ $, n \geq r$ (23)

And $TS(n) = \sum_{r=2}^{n} TS(n)_r$ (24)

These values constitutes the lower part (from the 4th column) of the binomial theorem coefficient, as it is shown in figure 4.

The use of generating functions to derive the above formula (23) and (24) (or relations (20) and (21) is now presented. Let $i_2(x)$ the generating function of the coefficient $TS(n)_2$:

$$i_2(x) = x^2(1 + 4x + 10x^2 + 20x^3 + \dots + \binom{n+1}{3}x^n + \dots) \quad (25)$$

$$i_2(x) = \sum_{n=2}^{\infty} \binom{n+1}{3} x^n \quad (26)$$

that is to say in a developed form:

$$i_2(x) = x^2 \sum_{i=0}^{\infty} \frac{(i+3)!}{3!i!} x^i \quad (27)$$

and dividing by 3! It comes:

$$i_2(x) = x^2\left[1+\sum_{i=0}^{\infty}\prod_{j=4}^{i+3} j\frac{x!}{i!}x^i\ \right] \tag{28}$$

and finally;

$$i_2(x) = x^2(1-x)^{-4} = \frac{x^2}{(1-x)^4} \tag{29}$$

A generalization of this result to separators with r output streams leads to:

$$i_3(x) = x^3(1-x)^{-5} = \frac{x^3}{(1-x)^5} \quad \text{for r =3} \tag{30}$$

$$i_r(x) = x^r(1-x)^{-r-2} = \frac{x^r}{(1-x)^{r+2}} \quad \text{for the general case} \tag{31}$$

Thus, the generating function of TS(n) is given by :

$$i\ (x) = x^2\sum_{r=2}^{\infty} i_r(x) \tag{32}$$

$$i\ (x) = \frac{x^2}{(1-x)^4}\left(1+\frac{x}{1-x}+\frac{x^2}{(1-x)^2}+....\right) \tag{33}$$

$$i\ (x) = \frac{x^2}{(1-x)^4}\left(\frac{1}{1-\dfrac{x}{1-x}}\right) \qquad -1/2 \le x \le 1/2 \tag{34}$$

$$i\ (x) = \frac{x^2}{(1-2x)(1-x)^3} \tag{35}$$

Table 9 shows the values of $TS(n)_2$, $TS(n)_3$ and TS(n) versus n; and the rate number of distinct separators to the total number of separators of all sequences. It can be noted that this rate drastically decreases when the number of components increases.

n	2	3	4	5	6	7
$TS(n)_2$	1	4	10	20	35	56
$TS(n)_3$	0	1	5	15	35	70
TS(n)	1	5	16	42	99	219
\f(number of distinct separators; total number of separators of all sequences	100%	100%	61%	30%	13%	5%

Table9: Number of distinct separators for an- n-component mixture

Conclusion

A non recurrent formulation of the number of sharp separation sequences involving complex separators (i.e. separators having more than two outputs) is derived in this paper. This derivation requires the numbering of all possible separation structures determined by solving equations in the space of integer numbers.

The use of generating functions is an elegant way to formulate the number of distinct separators sequences and the number of two-or-three output separator sequences for an n-component mixture. It can be observed that the number of distinct separators is always small; it is an interesting feature insofar as this number gives a lower bound on the total computational effort for scanning the tree of separation sequences.

Thus, it appears that the elucidation of the main combinatoric points will enable in the next future the solution of complex sharp separation problems by means of combinatorial optimization techniques, as for example the simulated annealing procedure.

Appendix 1: Determination of the coefficients of equations $n_s = P(n)$

Let $m_0 + 2m_1 + \dots + im_{i-1} + \dots + (k+1)m_k = n-1 (n \in N)$ an equation of the type (9), and n_s the number of integer solutions of a such an equation then, (G.Th.Guilbaud, 1990), n_s is given by :

$$n_s = a_{k,i}(n-1)^k + a_{k-1,i}(n-1)^{k-1} + \dots + a_{1,i}(n-1) + a_{0,i}$$

where $a_{j,i}$ are periodic coefficients, with period T=lowest common multiple of numbers 1,2,...,k+1. The determination of all $a_{j,i}$ (i=1 to T) coefficients is made by solving the T following systems:

$$\left|\begin{array}{l} a_{k,i}(n-1)^k + a_{k-1,i}(n-1)^{k-1} + \dots + a_{1,i}(n-1) + a_{0,i} = \alpha_{1,i} \\ a_{k,i}(n-1+T)^k + a_{k-1,i}(n-1+T)^{k-1} + \dots + a_{1,i}(n-1+KT) + a_{0,i} = \alpha_{k+1,i} \\ \dots\dots \qquad\qquad\qquad\qquad\qquad\qquad i = 1 to \quad T \\ a_{k,i}(n-1+KT)^k + a_{k-1,i}(n-1+KT)^{k-1} + \dots + a_{1,i}(n-1+KT) + a_{0,i} = \alpha_{k+1,i} \end{array}\right.$$

where $\alpha_{j,i}$ is the jth value ($1 \le j \le k+1$) of n_s for the ith period, i.e. the (ij)th value of n_s given, for example, in Tables 2 to 4. These systems are equivalent to:

$$\begin{pmatrix} 1 & n-1 & (n-1)^2 & \dots & (n-1)^k \\ 1 & n-1+T & (n-1+T)^2 & \dots & (n-1+T)^k \\ \dots & \dots & \dots & \dots & \dots \\ 1 & n-1+KT & (n-1+KT)^2 & \dots & (n-1+KT)^k \end{pmatrix} \begin{pmatrix} a_0 \\ i \\ a_1 \\ i \\ \dots \\ a_{k-1} \\ i \\ a_k \\ i \end{pmatrix} = \begin{pmatrix} \alpha 1 \\ i \\ \alpha_2 \\ i \\ \dots \\ \alpha_k \\ i \\ \alpha_{k+1} \\ i \end{pmatrix}, i=1 \text{ to } T$$

For solving these T systems, the T previous Van der Monde Matrices must be inverted. The method of solution (W.H. Press et al., 1988) is closely related to Lagrange's polynomial interpolation formula. Let $P_j(x)$ be the polynomial of degree k defined by:

$$P_j(x) = \prod_{l=0, l \neq j}^{k} \frac{x-(n-1+lT)}{(jT-lT)} = \sum_{l=1}^{k+1} A_{jl} \times x^{l-1} \qquad , j=1 \text{ to } k$$

Then the values of coefficients are:

$$A_{j,i} = \sum_{l=1}^{k+1} A_{lj}\alpha_{li} \qquad\qquad j=1 \text{ to } k,\ i=1 \text{ to } T$$

For example, let n_s the number of integer solutions of the equation:
$m_0 + 2m_1 + 3m_2 + 4m_3 = n-1$

Then,
$n_s = a_{3,i}(n-1)^3 + a_{2,i}(n-1)^2 + a_{1,i}(n-1) + a_{0,i}$

where $a_{j,i}$ are periodic coefficients, with period T = lowest common multiple of numbers 1,2,3,4 = 12. The determination of all $a_{j,i}$ (i=1 to 12) coefficients is made by the solution of the 12 following systems:

$$\left|\begin{array}{l} a_{3,i}(n-1)^3 + a_{2,i}(n-1)^2 + a_{1,i}(n-1) + a_{0,i} = \alpha_{1,i} \\ a_{3,i}(n-1+T)^3 + a_{2,i}(n-1+T)^2 + a_{1,i}(n-1+T) + a_{0,i} = \alpha 2, i \\ a_{3,i}(n-1+2T)^3 + a_{2,i}(n-1+2T)^2 + \ldots + a1, i(n-1+2T) + a_{0,i} = \alpha 3, i \\ a_{3,i}(n-1+3T)^3 + a_{2,i}((n-1+3T)^2 + \ldots + a_{1,i}(n-1+3T) + a_{0,i} = \alpha_{4,i} \end{array}\right. \qquad \text{i=1 to 12}$$

$$\text{where } \alpha = \begin{pmatrix} 1 & 1 & 2 & 3 & 5 & 6 & 9 & 11 & 15 & 18 & 23 & 27 \\ 34 & 39 & 47 & 54 & 64 & 72 & 84 & 94 & 108 & 120 & 136 & 150 \\ 169 & 185 & 206 & 225 & 249 & 270 & 297 & 321 & 351 & 378 & 411 & 441 \\ 478 & 511 & 551 & 588 & 632 & 672 & 720 & 764 & 816 & 864 & 920 & 972 \end{pmatrix}$$

the first line of the previous matrix is given in Table 4.
The solutions of the 12 Van der Monde systems leads to

$$a_{0,i} = \frac{(n-1+T)(n-1+2T)(n-1+3T)(n-1+3T}{6T^3}\alpha_{1,i} - \frac{(n-1)(n-1+2T)(n-1+3T)}{2T^3}\alpha_{2,i}$$
$$+ \frac{(n-1)(n-1+T)(n-1+3T}{2T^3}\alpha_{3,i} - \frac{(n-1)(n-1+2T)(n-1+3T)}{6T^3}\alpha_{4,i}$$

$$a_{1,i} = -\frac{(n-1+T)(n-1+2T)+(n-1+T)(n-1+3T)+(n-1+2T)(n-1+3T)}{6T^3}\alpha_{1,i} +$$
$$\frac{(n-1)(n-1+2T)+(n-1)(n-1+3T)+(n-1+2T)(n-1+3T)}{2T^3}\alpha_{2,i} -$$
$$\frac{(n-1)(n-1+T)+(n-1)(n-1+3T)+(n-1+T)(n-1+3T)}{2T^3}\alpha_{3,i} +$$
$$\frac{(n-1)(n-1+T)+(n-1)(n-1+2T)+(n-1+T)(n-1+2T)}{6T^3}\alpha_{4,i}$$

$$a_{2,i} = -\frac{(n-1+T)+(n-1+2T)+(n-1+3T)}{6T^3}\alpha_{1,i} - \frac{(n-1)+(n-1+2T)+(n-1+3T)}{2T^3}\alpha_{2,i}$$
$$+ \frac{(n-1)+(n-1+T)+(n-1+3T)}{2T^3}\alpha_{3,i} - \frac{(n-1)+(n-1+T)+(n-1+2T)}{6T^3}\alpha_{4,i}$$

$$a_{3,i} = -\frac{\alpha_{1i}}{6T^3} + \frac{\alpha_{2i}}{2T^3} - \frac{\alpha_{3i}}{2T^3}\alpha_{3,i} + \frac{\alpha_{4i}}{6T^3}\alpha_{4,i}$$

with T = 12

then the computation gives:

$$d = \begin{pmatrix} 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 & 1/144 \\ 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 & 5/48 \\ 1/2 & 7/16 & 1/2 & 7/16 & 1/2 & 7/16 & 1/2 & 7/16 & 1/2 & 7/16 & 1/2 & 7/16 \\ 1 & 65/144 & 76/177 & 81/144 & 128/144 & 49/144 & 108/144 & 65/144 & 112/144 & 81/144 & 92/144 & 49/144 \end{pmatrix}$$

So,

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{1}{2}(n-1) + 1 \qquad \text{if } n \equiv 1 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{7}{16}(n-1) + \frac{65}{144} \qquad \text{if } n \equiv 2 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{1}{2}(n-1) + \frac{76}{144} \qquad \text{if } n \equiv 3 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{7}{16}(n-1) + \frac{81}{144} \qquad \text{if } n \equiv 4 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{1}{2}(n-1) + \frac{128}{144} \qquad \text{if } n \equiv 5 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{7}{16}(n-1) + \frac{49}{144} \qquad \text{if } n \equiv 6 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{1}{2}(n-1) + \frac{108}{144} \qquad \text{if } n \equiv 7 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{7}{16}(n-1) + \frac{65}{144} \qquad \text{if } n \equiv 8 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{1}{2}(n-1) + \frac{112}{144} \qquad \text{if } n \equiv 9 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{7}{16}(n-1) + \frac{81}{144} \qquad \text{if } n \equiv 10 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{1}{2}(n-1) + \frac{92}{144} \qquad \text{if } n \equiv 11 \bmod 12$$

$$ns = \frac{1}{144}(n-1)^3 + \frac{5}{48}(n-1)^2 + \frac{7}{16}(n-1) + \frac{49}{144} \qquad \text{if } n \equiv 0 \bmod 12$$

Appendix 2: Derivation of relation (19)

This relation (19) is derived recursively.
For n = 2:

$$D_2(g(x)) = \frac{1}{2!} g''(x)(g(x)(4-3g(x)))^3$$

$$= 3g(x)-2$$

$$= \frac{1}{2} 2.(3g(x)-2) \quad \text{from (18)}$$

if, for n≥2, the relation (19) is true then:

$$D_{n+1}(g(x)) = \frac{(g(x)(4-3g(x)))^{2n+1}}{(n+1)!} g^{(n)}(x)$$

$$= \frac{(g(x)(4-3g(x)))^{2n+1}}{(n+1)!} \frac{d(\frac{n!}{g(x)(4-3g(x)))^{2n-1}} D_n(g(x)))}{dx}$$

$$= \frac{(g(x)(4-3g(x)))^{2n+1}}{(n+1)} \frac{d(\frac{D_n(g(x))}{g(x)(4-3g(x)))^{2n-1}})}{dx}$$

$$= \frac{(g(x)(4-3g(x)))^{2n+1}}{(n+1)}$$

$$\left[\frac{D'_n(g(x)).(g(x)(4-3g(x)))^{2n-1}}{(g(x)(4-3g(x)))^{4n-2}} - \frac{D_n(g(x)).(2n-1).(4g'(x)-6g(x)g'(x)).(g(x)(4-3g(x)))^{2n-2}}{(g(x)(4-3g(x)))^{4n-2}} \right]$$

$$= \frac{D'_n(g(x))(g(x)(4-3g(x))^2 + 2(2n-1)D_n(g(x))(3g(x)-2))}{n+1}$$

because $g'(x) = (g(x)(4-3g(x)))^{-1}$.
Then, the relation (19) is also true for n+1

Appendix 3: Derivation of equivalence of relations (20) and (22)

This equivalence is derived recursively.

For n=2:

$TS(n)_2 = 1$ from (22)

$TS(n)_2 = \binom{3}{3} = 1$ from (20)

If, for $n \geq 2$, relations (20) and (22) are equivalent then:

$$TS(n)_2 = \sum_{i=1}^{n-1} i(n-i) = \binom{n+1}{3} = \frac{n^3 - n}{6}$$

$$TS(n+1)_2 = \sum_{i=1}^{n} i(n+1-i) = \sum_{i=1}^{n} [i(n-i) + i]$$

$$= TS(n)_2 + \frac{1}{2} n(n+1)$$

$$= \frac{(n-1)n(n+1)}{6} + \frac{3n(n+1)}{6}$$

$$= \frac{n(n+1)(n+2)}{6}$$ that proves the equivalence for n+1.

The derivation of the other relations (for $TS(n)_3$, ..., $TS(n)_r$) is made in the same way

Notation

$g(x)$	=	generation function of number of sequences S_n
$i_r(x)$	=	generation function of number of distinct separators $TS(n)_r$
I_n	=	set of admissible structure of sharp separators
m	=	total number of separators in a sequences = $\sum_{m_k \in I_n} m_k$
m_k	=	number of separators with one input and (k+2) outputs
n	=	number of components to be separated
n_s	=	number of distinct structures = number of elements of I_n
S_n	=	number of separation sequences of an n-component mixture
$TS(n)_r$	=	number of distinct separators with r output streams, for an n-component separation
$TS(n)$	=	total number of distinct separators for sequences of separation of n-component mixture

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