JOURNAL OF CHEMOMETRICS
J. Chemometrics 2002; **16**: 609–612
Published online in Wiley InterScience (www.interscience.wiley.com). DOI: 10.1002/cem.760

Non-triviality and identification of a constrained Tucker3 analysis

Jos M. F. ten Berge[1] and Age K. Smilde[2]*

[1]Heijmans Institute, University of Groningen, Grote Kruisstraat 2/1, NL-9712 TS Groningen, The Netherlands
[2]Process Analysis and Chemometrics, Department of Chemical Engineering, University of Amsterdam, Nieuwe Achtergracht 166, NL-1018 WV Amsterdam, The Netherlands

Received 24 October 2001; Accepted 30 May 2002

Properties of estimated parameters of models of chemical systems are important. This paper focuses on two properties of such estimated parameters: triviality and uniqueness. If a chemical system is analyzed using a Tucker3 model, then the resulting core can often be rotated to a simple structure containing zeros. This means that it is possible that a prespecified pattern of zero and non-zero elements of the core, as used in e.g. constrained Tucker3 models, is not an active constraint; that is, the zeros can be obtained trivially for free. Once a non-trivial pattern of zeros in the core is specified, the question arises whether this pattern is sufficient for obtaining unique loadings. Both issues are discussed in this paper and it is shown that the model used by Gurden *et al.* (*J. Chemometrics* 2001; 15: 101–121) does essentially involve a non-trivial core and implies rotationally unique parameter estimates. Copyright © 2002 John Wiley & Sons, Ltd.

KEYWORDS: Candecomp/Parafac; Tucker3; uniqueness; grey models; constrained Tucker; typical rank

1. INTRODUCTION

One of the leading themes in chemometrics is uniqueness of solutions obtained by resolving chemical mixtures or by modeling chemical systems in general. In the class of problems known as curve resolution, where chemical mixtures are resolved into pure component spectra and profiles, uniqueness plays a dominant role [1,2]. The reason for the importance of uniqueness of solutions is clear: obtaining unique solutions allows for an unambiguous interpretation of the estimated parameters (spectra, chromatograms, etc.) in terms of the chemical system. In short, uniqueness generates fundamental insight into the chemistry of the studied system.

The popularity of the Candecomp/Parafac (CP) model in chemistry is due to its uniqueness properties [3,4]. This makes the CP model suitable for performing second-order calibration [5,6], for resolving mixtures using fluorescence spectroscopy [7] and for studying chemical processes [8].

In contrast with CP, rotational indeterminacy is one of the distinguishing features of Tucker3 PCA [9] of three-way arrays. After fitting the model by Tuckals3 [10], there is freedom of (obliquely) rotating the core in three directions under the proviso that the inverse rotations are applied to all three component matrices involved. However, when certain elements of the core are constrained to be zero (thereby obtaining a constrained or restricted Tucker3 model), the rotational indeterminacy is confined, sometimes to the point that the solution becomes unique up to permutations and scaling; that is, the identification of the model reaches the same level as that of Candecomp/Parafac [3,4]. An example to that effect has been given involving a $3 \times 3 \times 3$ core with 18 elements zero [11]. In chemistry a restricted Tucker3 model has been used to calibrate a second-order sensor [12]. Later it was shown that the specific pattern of zero-constrained core elements gave partial uniqueness, allowing for the unique determination of analyte concentrations [13].

Apart from the identification issue, there is the issue of triviality. The very rotational freedom of a Tuckals3 solution is well known to imply a variety of patterns of zero elements that can be attained trivially; for a review, see Reference [14]. Questions of triviality (is the 'model' truly a model or is it an artifact?) and uniqueness (is the model identified?) arise whenever cores are constrained to have a vast majority of zero elements.

A particular case of interest, to be examined in this paper, is a core appearing in Reference [15]. Gurden *et al.* [15] used a

*Correspondence to: A. K. Smilde, Process Analysis and Chemometrics, Department of Chemical Engineering, University of Amsterdam, Nieuwe Achtergracht 166, NL-1018 WV Amsterdam, The Netherlands.
E-mail: asmilde@science.uva.nl

constrained core, rewritten in terms of horizontal slices, of the form

$$\mathbf{G}_a = [\mathbf{G}_1|\mathbf{G}_2|\mathbf{G}_3|\mathbf{G}_4|\mathbf{G}_5] =$$

$$\left[\begin{array}{ccccc|ccccc|ccccc|ccccc|ccccc} g_{111} & 0 & 0 & 0 & 0 & 0 & g_{122} & 0 & 0 & 0 & 0 & 0 & g_{133} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & g_{244} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & g_{355} \end{array}\right] \quad (1)$$

This model, christened 'grey model', can be examined in terms of non-triviality and uniqueness. In the next sections it will be shown that models based on a core like (1) are non-trivial and partly unique. It will also be explained that additional constraints employed by Gurden *et al.* remove precisely the partial indeterminacy implied by the core constraints, whence their model is fully identified. This opens the door for unambiguously interpreting the estimated loadings in terms of the chemical process under study and gives fundamental insight into the nature of the variation found in this chemical process.

2. NON-TRIVIALITY OF THE MODEL

The question of triviality of (1) can be dealt with by considering the three-way rank of $\underline{\mathbf{G}}$. It is obvious that the slices of $\mathbf{G}_a$ contain at least five linearly independent rows. Therefore a full decomposition by CP will need at least five components. Because the number of components required to get a perfect CP fit coincides with the three-way rank of any array [16], it can be inferred that the array has at least rank 5. On the other hand, the array also has at most rank 5. This is immediate from the consideration that only five elements are non-zero, each of which can be accounted for by a three-way array of rank 1. It can be concluded that the core array $\underline{\mathbf{G}}$ above has rank 5.

To show that rank 5 is usually not possible, we shall resort to the concept of typical rank. The typical rank of an array is the rank the array has with positive probability when the elements of the array are sampled randomly from a continuous distribution. For a matrix the typical rank (the rank we observe almost surely) is also the maximal rank. For instance, a $p \times q$ matrix, $p > q$, will have rank q when it is sampled randomly from a continuous distribution. For three-way arrays, however, maximal rank does not coincide with typical rank. For example, a $2 \times 2 \times 2$ array can have rank 0, 1, 2 or 3. If the eight elements of the $2 \times 2 \times 2$ array are filled randomly with numbers drawn from a normal distribution, then in 79% of the cases the array has rank 2 and in 21% of the cases the array has rank 3. This means that rank 0 and rank 1 arrays occur with probability zero [17]. The typical rank of a $2 \times 2 \times 2$ array is then 2 or 3 (i.e. the set {2,3}). Another example is the case of $7 \times 3 \times 3$ arrays. Their maximal rank is 8 and the typical rank is 7 [18]. This means that rank 8 will never be observed after random sampling from a continuous distribution.

To arrive at non-triviality of (1), we need to establish that $5 \times 5 \times 3$ arrays of rank 5 arise with probability zero. Unfortunately, the few general rules available to determine the typical rank of three-way arrays [18] do not cover $5 \times 5 \times 3$ arrays. However, we can establish the following lower bound to the typical rank of such arrays.

Result

The typical rank of $p \times p \times 3$ arrays is at least $p + 1$.

Proof

Suppose a $p \times p \times 3$ array $\underline{\mathbf{X}}$ has rank less than p, and let a matricized form of $\underline{\mathbf{X}}$ be the $p \times 3p$ matrix $\mathbf{X}$. Then $\mathbf{X}\mathbf{X}^{\mathrm{T}}$ has determinant zero. Since this determinant is an analytic function of the elements of $\mathbf{X}$ and is not zero everywhere (for every array $\underline{\mathbf{X}}$), it follows that the determinant is almost surely non-zero; see Reference [19], Theorem 5.A.2. It remains to investigate the possibility of having $\underline{\mathbf{X}}$ of rank p.

When $\underline{\mathbf{X}}$ has rank p, and $\mathbf{X}_1$ is non-singular, the three $p \times p$ slices $\mathbf{X}_1$, $\mathbf{X}_2$ and $\mathbf{X}_3$ can be decomposed by CP as

$$\mathbf{X}_k = \mathbf{A}\mathbf{C}_k\mathbf{B}^{\mathrm{T}} \quad (2)$$

with $\mathbf{A}$ and $\mathbf{B}$ non-singular and $\mathbf{C}_k$ a diagonal matrix, $k = 1, 2, 3$. It follows that $\mathbf{X}_2\mathbf{X}_1^{-1} = \mathbf{A}\mathbf{C}_2\mathbf{C}_1^{-1}\mathbf{A}^{-1}$ and $\mathbf{X}_3\mathbf{X}_1^{-1} = \mathbf{A}\mathbf{C}_3\mathbf{C}_1^{-1}\mathbf{A}^{-1}$. Hence $\mathbf{X}_2\mathbf{X}_1^{-1}$ and $\mathbf{X}_3\mathbf{X}_1^{-1}$ commute, which means that

$$\|\mathbf{X}_2\mathbf{X}_1^{-1}\mathbf{X}_3\mathbf{X}_1^{-1} - \mathbf{X}_3\mathbf{X}_1^{-1}\mathbf{X}_2\mathbf{X}_1^{-1}\|^2 = 0 \quad (3)$$

The set of arrays $\underline{\mathbf{X}}$ such that $\mathbf{X}_1$ is singular is closed. Therefore the complement set is open. The sum of squares (3) is an analytic function of the elements of $\underline{\mathbf{X}}$ defined on that complement set. Because the function is not the zero function, it is non-zero almost surely [19] on the same complement set. The fact that (3) will take the value zero when the rank of $\underline{\mathbf{X}}$ is p, combined with the fact that $\mathbf{X}_1$ is non-singular almost surely, shows that $\underline{\mathbf{X}}$ has rank p with probability zero. It has thus been proven that a $p \times p \times 3$ array has rank higher than p almost surely.

Specifically, the result implies that a $5 \times 5 \times 3$ array has rank 6 or above almost surely. Hence ranks lower than 6 arise with probability zero when the array is randomly sampled from a continuous distribution. It may be objected that this assumption is not met when a core array arises at convergence of the unconstrained Tuckals3 procedure. Indeed, this procedure might have a tendency to produce unusual non-random results. However, practical experience with Tuckals3 has revealed no such tendency at all. Tuckals3 cores seem to behave as if randomly sampled from a continuous distribution. This justifies the inference that the typical rank of $5 \times 5 \times 3$ Tuckals3 core arrays is at least 6. Because rotation of an array in any direction does not change the array rank, it follows that the pattern of (1) cannot be

obtained trivially by rotating any obtained core afterwards in three directions. The Gurden *et al.* core array thus represents a model rather than an artifact.

3. PARTIAL UNIQUENESS OF THE MODEL

In the model of Gurden *et al.* the three modes consist of different batches (with $I \times 3$ loading matrix **A**), different wavelengths (with $J \times 5$ loading matrix **B**) and different points in time (with $K \times 5$ loading matrix **C**). The uniqueness of the model pertains to the question of whether or not non-singular matrices **S** (3×3), **T** (5×5) and **U** (5×5) can be found, other than rescaled permutation matrices, such that the core $\mathbf{SG}_a(\mathbf{U}^T \otimes \mathbf{T}^T)$ has zeros in the same places as $\mathbf{G}_a$ of (1). Such transformations are allowed provided that the loading matrices **A, B** and **C** are transformed to $\mathbf{AS}^{-1}$, $\mathbf{BT}^{-1}$ and $\mathbf{CU}^{-1}$ respectively. In fact, this is partly the case. As will be shown below, the matrix **S** is unique (up to permutations and scaling) and so are the last two rows and last two columns of **T** and **U**. There is, however, non-uniqueness in the remaining parts of **T** and **U**; that is, **T** can have an arbitrary 3×3 submatrix in its upper left-hand corner, compensated for by having its inverse as corresponding submatrix in **S**.

Proof

To investigate the uniqueness properties of the array (1), it is convenient to rearrange it into an array containing the horizontal slices. Thus, instead of considering the uniqueness of $\mathbf{G}_a$, we shall consider the equivalent uniqueness properties of

$$\mathbf{G}_h = [\mathbf{H}_1|\mathbf{H}_2|\mathbf{H}_3] = \left[\begin{array}{ccccc|ccccc|ccccc} g_{111} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & g_{122} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & g_{133} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & g_{244} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & g_{355} \end{array}\right] \tag{4}$$

which has a transformed version $\mathbf{TG}_h(\mathbf{S}^T \otimes \mathbf{U}^T)$. Without loss of generality, let the non-zero elements of $\mathbf{G}_h$ be scaled to unity.

We start with the matrix $\mathbf{S}^T$, producing linear combinations of the horizontal slices $\mathbf{H}_1$, $\mathbf{H}_2$ and $\mathbf{H}_3$. If the new slices are to have zeros in the same places as the old ones, it is obvious that they must have ranks 3, 1 and 1 respectively. Because the ranks of the new slices are independent of the non-singular matrices **T** and **U**, it is clear that **S** must preserve the rank pattern of $\mathbf{H}_1$, $\mathbf{H}_2$ and $\mathbf{H}_3$. Trivial mixes (by taking $\mathbf{S}^T = \mathbf{I}_3$, up to permutations and rescaling of its columns) will indeed preserve the ranks 3, 1 and 1. Non-trivial mixes, on the other hand, will produce ranks 2, 4 or 5, none of which are desired. It follows that $\mathbf{S}^T$ has to be $\mathbf{I}_3$, up to permutations and scaling. Without loss of generality, we shall set $\mathbf{S} = \mathbf{I}_3$.

Having established the essential uniqueness of **S**, it can be seen that $\widetilde{\mathbf{H}}_k = \mathbf{TH}_k\mathbf{U}^T$ must have zeros in the same places as $\mathbf{H}_k$, $k = 1, 2, 3$. Partition **T** and **U** into

$$\mathbf{T} = \begin{bmatrix} \mathbf{T}_{11} & \mathbf{T}_{12} \\ \mathbf{T}_{21} & \mathbf{T}_{22} \end{bmatrix}, \qquad \mathbf{U} = \begin{bmatrix} \mathbf{U}_{11} & \mathbf{U}_{12} \\ \mathbf{U}_{21} & \mathbf{U}_{22} \end{bmatrix} \tag{5}$$

with $\mathbf{T}_{11}$ and $\mathbf{U}_{11}$ of order 3×3, $\mathbf{T}_{12}$ and $\mathbf{U}_{12}$ of order 3×2, $\mathbf{T}_{21}$ and $\mathbf{U}_{21}$ of order 2×3 and $\mathbf{T}_{22}$ and $\mathbf{U}_{22}$ of order 2×2. We start with the rank 1 matrix $\mathbf{H}_3$. Writing $\widetilde{\mathbf{H}}_3 = \mathbf{TH}_3\mathbf{U}^T = \mathbf{Te}_5\mathbf{e}_5^T\mathbf{U}^T$, with $\mathbf{e}_5$ the fifth column of $\mathbf{I}_5$, we have $\widetilde{\mathbf{H}}_3 = \mathbf{t}_5\mathbf{u}_5^T$, with $\mathbf{t}_5$ and $\mathbf{u}_5$ the fifth columns of **T** and **U** respectively. Because $\widetilde{\mathbf{H}}_3$ has zeros in the same places as $\mathbf{H}_3$, both $\mathbf{t}_5$ and $\mathbf{u}_5$ have to be proportional to $\mathbf{e}_5$. A similar argument applied to $\widetilde{\mathbf{H}}_2$ shows that the fourth columns of **T** and **U** must be proportional to $\mathbf{e}_4$. It should be noted that this implies that $\mathbf{T}_{12}$ and $\mathbf{U}_{12}$ have all elements zero.

Finally, consider $\widetilde{\mathbf{H}}_1$. To prevent **T** and **U** from being singular, both $\mathbf{T}_{11}$ and $\mathbf{U}_{11}$ must be non-singular. Because submatrices $\mathbf{H}_{12}$ and $\mathbf{H}_{21}$ of $\mathbf{H}_1$, defined analogously to submatrices of **T**, have all elements zero, it follows that $\mathbf{T}_{21}$ and $\mathbf{U}_{21}$ are zero. It remains to consider $\mathbf{T}_{11}$ and $\mathbf{U}_{11}$. Clearly, taking any non-singular 3×3 matrix for $\mathbf{T}_{11}$ is allowed provided that $\mathbf{U}_{11}^T$ is its inverse. This pinpoints where the model is non-unique: there is freedom of rotation for the first three columns of the matrices **B** and **C** of the Tucker3 solution, albeit that the latter rotation must be the inverse of the former. All other parameters of **S, T** and **U** are unique up to joint permutations and scaling.

4. ADDITIONAL CONSTRAINTS USED BY GURDEN *ET AL.*

One may wonder what implications the above results have on the analysis of Gurden *et al.* [15] in particular. The 'grey model' they employed appears to have rotational indeterminacy for the 'white part' of the core array, which corresponds to the first three columns of **B** and **C**. Loosely stated, they still have the curve resolution problem of rotational ambiguity in resolving the pure component spectra and kinetic profiles. The variation in the first mode (the batch mode) is insufficient to remove this rotational freedom. However, they fixed the loadings of **B** (spectra) to *a priori* known spectra and this solved the problem of rotational freedom trivially, because this fixes $\mathbf{T}_{11}$ up to scaling, and hence $\mathbf{U}_{11}$. Thus the Gurden *et al.* model is fully identified. Note that the uniqueness of **A** ($\mathbf{S} = \mathbf{I}_3$) justifies the interpretation made by Gurden *et al.* of the first loading vector in **A** as reflecting the differences in initial concentration of the reactant between batches.

Another way of identifying the model is by imposing a first-order kinetic model on the profiles in the time mode (mode C). This is also sufficient for removing the rotational ambiguity in the 'white part' of the model [20]. By imposing the kinetic model on the C mode, kinetic constants can be calculated and pure spectra can be obtained [21].

5. DISCUSSION

Preferably, non-triviality and uniqueness of core-con-

strained Tucker3 models should be determined on the basis of general rules. Research in this area is quite complicated and progress has been slow. General principles about uniqueness are totally absent, which means that, for the time being, we have to resort to *ad hoc* arguments. For the determination of non-triviality, however, the prospects are better. Non-triviality can be based on arguments of typical rank or maximal simplicity (the largest number of zeros that arrays of any particular order usually can have). Typical rank results of some generality are available [18], although the particular case examined in this paper did require an *ad hoc* extension. A few maximal simplicity rules can be found in Reference [14].

6. CONCLUSIONS

Uniqueness of solutions of parameter estimates of models for chemical systems is important, since it allows for an unambiguous interpretation of the found estimates. It has been shown that the model used by Gurden *et al.* [15] gives unique solutions.

Acknowledgement

We thank Tom Snijders (Department of Sociology, University of Groningen) for useful comments.

REFERENCES

1. Manne R. On the resolution problem in hyphenated chromatography. *Chemometrics Intell. Lab. Syst.* 1995; **27**: 89–94.
2. Tauler R, Smilde AK and Kowalski BR. Selectivity, local rank, three-way data analysis and ambiguity in multivariate curve resolution. *J. Chemometrics* 1995; **9**: 31–58.
3. Carroll JD and Chang J. Analysis of individual differences in multidimensional scaling via an *N*-way generalization of 'Eckart–Young' decomposition. *Psychometrika* 1970; **35**: 283–319.
4. Harshman RA. Foundations of the PARAFAC procedure: models and conditions for an 'explanatory' multimodal factor analysis. *UCLA Working Papers Phonet.* 1970; **16**: 1–84.
5. Kiers HAL and Smilde AK. Some theoretical results on second-order calibration methods for data with and without rank overlap. *J. Chemometrics* 1995; **9**: 179–195.
6. Sanchez E and Kowalski BR. Tensorial calibration: 2. Second-order calibration. *J. Chemometrics* 1988; **2**: 265–280.
7. Ross RT, Lee C, Davis CM, Ezzeddine BM, Fayyad EA and Leurgans SE. Resolution of the fluorescence spectra of plant pigment-complexes using trilinear models. *Biochim. Biophys. Acta* 1991; **1056**: 317–320.
8. Bro R. Exploratory study of sugar production using fluorescence spectroscopy and multi-way analysis. *Chemometrics Intell. Lab. Syst.* 1999; **46**: 133–147.
9. Tucker LR. Some mathematical notes on three-mode factor analysis. *Psychometrika* 1966; **31**: 279–311.
10. Kroonenberg PM and De Leeuw J. Principal component analysis of three-mode data by means of alternating least squares algorithms. *Psychometrika* 1980; **45**: 69–97.
11. Kiers HAL, ten Berge JMF and Rocci R. Uniqueness of three-mode factor models with sparse cores: the $3 \times 3 \times 3$ case. *Psychometrika* 1997; **62**: 349–374.
12. Smilde AK, Tauler R, Henshaw JM, Burgess LW and Kowalski BR. Multicomponent determination of chlorinated hydrocarbons using a reaction-based chemical sensor. 3. Medium-rank second-order calibration with restricted Tucker models. *Anal. Chem.* 1994; **66**: 3345–3351.
13. Kiers HAL and Smilde AK. Constrained three-mode factor analysis as a tool for parameter estimation with second-order instrumental data. *J. Chemometrics* 1998; **12**: 125–147.
14. Rocci R and ten Berge JMF. Transforming three-way arrays to maximal simplicity. *Psychometrika* 2002; **67**(3): 351–365.
15. Gurden SP, Westerhuis JA and Smilde AK. Modelling of spectroscopic batch process data using grey models to incorporate external information. *J. Chemometrics* 2001; **15**: 101–121.
16. Kruskal JB. Three-way arrays: rank and uniqueness of trilinear decompositions with applications to arithmetic complexity and statistics. *Linear Algebra Appl.* 1977; **18**: 95–138.
17. Kruskal JB. Rank, decomposition, and uniqueness for 3-way and *N*-way arrays. In *Multiway Data Analysis*, Coppi R, Bolasco S (eds). Elsevier: Amsterdam, 1989; 8–18.
18. Ten Berge JMF. The typical rank of tall three-way arrays. *Psychometrika* 2000; **65**: 525–532.
19. Fisher RM. *The Identification Problem in Econometrics*. McGraw-Hill: New York, 1966.
20. Smilde AK, Hoefsloot HCJ, Kiers HAL, Bijlsma S and Boelens HFM. Sufficient conditions for unique solutions within a certain class of curve resolution models. *J. Chemometrics* 2001; **15**: 405–411.
21. Gurden SP, Westerhuis JA and Smilde AK. Monitoring of batch processes using spectroscopy. *AIChE J.* 2002; **48**: 2283–2297.

