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Application of Derringer's desirability function for the selection of optimum separation conditions in capillary zone electrophoresis

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Abstract

In order to obtain a set of optimal experimental conditions for the separation of rare earth metal ions in capillary electrophoresis (CE), a multicriteria approach is applied. For this purpose, the Derringer's desirability function is proposed to determine conditions that will result in the most desirable combination of separation, sensitivity and analysis time. The CE separation method was optimized with the aid of a central composite design for two variables, i.e. the pH and the concentration of a complexing agent in the buffer electrolyte.

Keywords: Derringer's desirability function; Central composite designs; Multicriteria decision making; Optimization; Chemometrics; Rare earth ions; Metal ions

1. Introduction

Due to its extremely high separation efficiency, capillary electrophoresis (CE) of low-molecular-mass ions has proven to be a powerful tool for the analysis of complex mixtures of ions [1–6]. An interesting complex mixture is the group of rare earth (RE) metal ions. Recently, a CE method was optimized for the separation of four REs [6]. The separation was enhanced by the addition of α -hydroxyisobutyric acid (HIBA) as a chelating agent to increase the difference in electrophoretic mobility between the REs. For the optimization, a central composite experimental design for two variables, i.e. the pH and the concentration of HIBA was applied.

In separation techniques, one not only has to consider separation, but other optimization goals,

such as sensitivity, peak shape and analysis time, as well. Often one or more of the criteria which are being studied are in conflict with each other regarding the optimum. It is well known that in high-performance liquid chromatography (HPLC) the pay-off for a good separation often is a long analysis time [7,8]. In CE, this can also be the case. Additionally, one also has to pay more attention to the sensitivity, which is a general problem in CE and which can decrease with increasing separation. In order to find the best compromise between conflicting criteria, the selection of the optimal conditions therefore requires a multicriteria decision making (MCDM) approach.

Various MCDM procedures were described and applied in experimental chemistry [7–17]. A number of these appear to be useful for bicriteria optimizations. As an example, the utility functions, the threshold approach and the concept of Pareto op-

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tinality can be mentioned [8]. The theory behind these procedures is described in Refs. [9–11]. In optimization procedures where more than three criteria are being studied, MCDM techniques such as PROMETHEE [12], desirability functions [8,13,14] and the target vector criterion [15,16] were shown to be appropriate.

In the present study it is intended to investigate the usefulness of Derringer's desirability functions in tackling a CE optimization procedure with a multi-criteria (MC) problem. Derringer's desirability function was introduced in HPLC by Bourguignon and Massart [8] for the selection of the best chromatogram out of a number of possible chromatograms, and appeared to be suitable for solving this MC problem. As the final result of a CE analysis very much resembles that of HPLC, Derringer's desirability function is applied on the data obtained in the optimization of the separation of REs published in [6], where the separation (separation coefficient α), sensitivity (peak height H) and analysis time (t_m) were studied as responses. The optimum separation conditions in Ref. [6] were obtained by overlaying the response surfaces of the criteria. This can be considered as a kind of threshold procedure which was applied manually, and was found to be time-consuming. Because MCDM with the aid of Derringer's desirability function can be applied very easily using an adequate spreadsheet and because of our previous experience in HPLC, this procedure was selected and is applied.

The theory behind Derringer's desirability function is described extensively in Refs. [8,13,14]. In short, the measured properties of each criterion are transformed to a dimensionless desirability (d) scale. This makes it possible to combine results obtained for properties measured on different scales. The scale of the desirability function ranges between $d=0$, for a completely undesirable response, to $d=1$ for a fully desired response, above which further improvements would have no importance. There are two types of transformations possible, a one-sided and a two-sided transformation. The one-sided transformation is applied to cases where one has to minimise or maximise a response. For the situations of resolution, sensitivity, etc., in which one wants to maximise the response, Eq. (1) is applied (one-sided transformation). This results in functions as shown in Fig. 1a.

$$d_i = 0 \quad \text{if } Y_i \leq Y^-$$

$$d_i = \left(\frac{Y_i - Y^-}{Y^+ - Y^-} \right)^r \quad \text{if } Y^- < Y_i < Y^+ \quad (1)$$

$$d_i = 1 \quad \text{if } Y_i \geq Y^+$$

If the value of a criterion (Y_i) is equal to, or below, the lowest possible limit set (Y^-) for that criterion, then $d_i=0$. If Y_i is higher than, or equal to, the highest possible limit set (Y^+) for that criterion, then $d_i=1$.

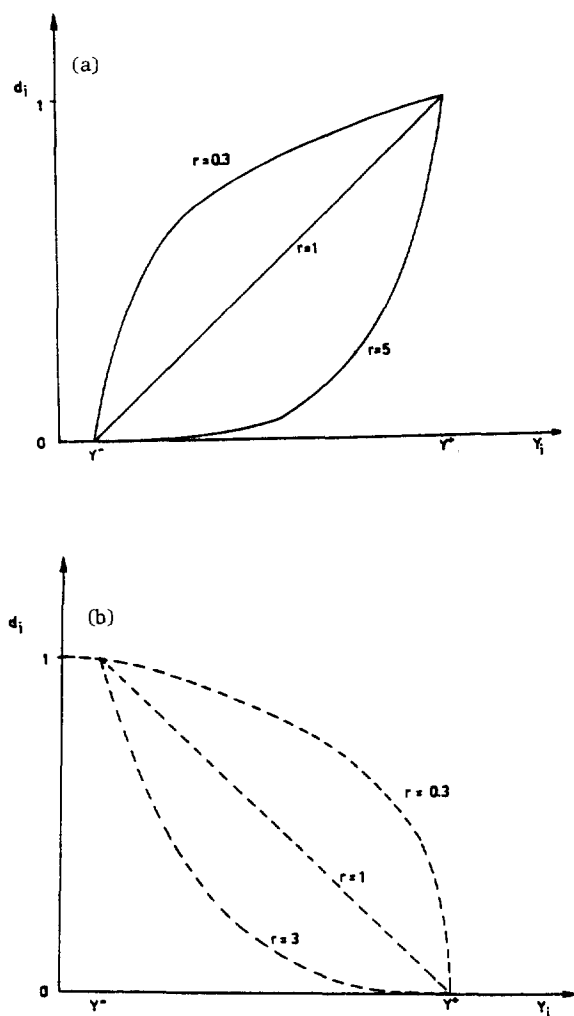


Fig. 1. Derringer's desirability functions; (a) the one-sided transformation for maximisation of a response variable; (b) the "reversed" one-sided transformation for minimisation of a response variable.

If Y_i is between the lowest and the highest level, then one takes the ratio of the difference between Y_i and Y^- to the difference between Y^+ and Y^- , powered by a factor, r , which describes the shape of the desirability function. As can be observed in Fig. 1 the factor r offers the user flexibility in the definition of the desirability function [8,14]. The value of the r factor is decided by the user and depends on the degree of desirability that one wants for a certain response between Y^+ and Y^- . For example, if one would like to obtain a degree of desirability of 0.5 for a response Y_i that corresponds with $0.5 \cdot Y^+$, representing a linear transformation function, then r can be calculated by substitution of these values into Eq. (1). This results in an r factor of 1, corresponding to a straight line (Fig. 1).

When minimisation of a response is required (analysis time, etc.), the opposite of these if/then rules (corresponding with Fig. 1b) are applied. For convenience, we will refer to this procedure as the “reversed” one-sided transformation. After transforming each criterion, the overall quality is calculated by determining the geometric mean (D) of the transformed values of the individual criteria. The value of D is the highest at conditions where the combination of the different criteria is globally optimal.

2. Experimental

The experimental conditions and the materials used are described in detail in Ref. [6]. The analysis was carried out on a Waters Quanta-4000 capillary electrophoresis system, equipped with a positive power supply. The capillaries were ordinary fused-silica capillaries (Waters AccuSep, 60 cm capillaries) with $75 \mu\text{m}$ I.D., and 52 cm length from the point of sample introduction to the point of detection. The electrophoresis zones were detected with a fixed-wavelength UV detector at 214 nm.

Stock solutions of 1000 mg l^{-1} of lanthanum and cerium were prepared in Milli-Q water using the nitrate salt of these ions. Terbium oxide and gadolinium oxide were first dissolved in an excess of ultra-pure nitric acid and evaporated to dryness. The residue was redissolved in Milli-Q water to obtain a 1000 mg l^{-1} stock solution. Standard solutions of the

REs were prepared daily. The buffer electrolyte consisted of a 10 mM creatinine–acetate buffer and various concentrations of HIBA. A 10 mg l^{-1} mixture of the REs was injected for 20 s by electromigration at 30 kV and the running voltage was 30 kV ($\pm 20 \mu\text{A}$).

3. Results and discussion

3.1. Separation described by the separation coefficient

The four RE metal ions for which the separation was optimized, were lanthanum, cerium, gadolinium and terbium. Two parameters have an important effect on this separation, i.e. the concentration of HIBA and the pH. These two parameters were examined with the aid of a central composite design (Table 1), in order to obtain the optimum separation conditions for this mixture [6]. The electropherograms corresponding to runs 1, 5, 7 and 8 (in Table 1) are shown in Fig. 2. In Ref. [6] not only the separation, but also the analysis time and the sensitivity were taken into account. Because the migration time cannot be modelled directly in CE studies, due to the possible changes in electroosmotic flow (EOF), in the previous report [6] the separation was quantified by the separation coefficient or separation factor (α) instead of the resolution (R_s). Direct modelling of the separation factor was possible in this specific case, as there was no crossing-over of the RE. The peak height (H), was used as a measure of the sensitivity. The analysis time (t_m) was expressed by means of the migration time of the last peak (migration time of Tb).

The applied model was a second order polynomial:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_{11}X_1^2 + b_{22}X_2^2 + b_{12}X_1X_2$$

($X_1 = \text{pH}$ and $X_2 = \text{HIBA}$) (2)

The regression coefficients were calculated using multiple regression analysis. The three separation coefficients ($\alpha_1 = t_m\text{Ce}/t_m\text{La}$, $\alpha_2 = t_m\text{Gd}/t_m\text{Ce}$ and $\alpha_3 = t_m\text{Tb}/t_m\text{Gd}$) were calculated and modelled. The same modelling procedure was performed for the

Table 1

The central composite experimental design and the obtained D values for each run and the selected optimum conditions

Run	pH	[HIBA]	$d-H_{\min}^1$	$d-t_m^2$	$d-\alpha_{\min}^3$	$d-R_{\min}^4$	$D-\alpha_{\min}^5$	$D-R_{\min}^6$
	3.70	2.00	0.90	0.72	0.28	0.14	0.57	0.45
	4.30	2.00	0.21	0.80	0.53	1	0.45	0.55
	3.70	8.00	0	0	1	1	0	0
	4.30	8.00	0	0.61	0.88	1	0	0
	3.58	5.00	0.33	0.42	0.67	1	0.45	0.52
	4.42	5.00	0	0	0.93	1	0	0
	4.00	0.74	0.63	0.84	0	0	0	0
	4.00	9.24	0	0	1	1	0	0
	4.00	5.00	0.29	0	0.95	1	0	0
Optimum	4.10	2.00	0.78	0.74	0.52	1	0.67	0.83

^{1,2,3} and ⁴ represent the desirability values for the minimum peak height, the analysis time, the minimum separation factor and the minimum resolution, respectively, obtained for each run.

⁵ and ⁶ represent the overall desirability calculated for the runs.

peak heights of the four REs and for the migration time of the last peak (t_m Tb). From the minimum separation coefficients (α_{\min}), the peak heights and the analysis time, optimal separation conditions for the four REs were derived by overlaying all the calculated response surfaces of the different criteria. Taking all the optimal regions of the criteria into account, the optimum was situated around pH 4.10 and with [HIBA]=2 mM. An electropherogram obtained at these conditions is shown in Fig. 3. As can be seen, a good separation is obtained within 3.5 min.

In contrast with the approach proposed by Bourguignon and Massart [8], in the current study it is intended to use the Derringer's functions for the transformation of the responses obtained for the included criteria into a desirability scale, in order to select the optimal conditions according to the overall quality response surface. Therefore, Derringer's transformation functions are applied to the values of the same criteria as before [6].

After modelling the peak height of all the peaks separately, the minimum peak height (H_{\min}) obtained for all the peaks over the whole experimental domain was selected and transformed to the d -scale. As mentioned before, the migration times cannot be used directly. Therefore, the effective mobilities of the ions and the mobility of the EOF were first modelled separately. With the mobilities of the ions and the mobility of the EOF, one can calculate the migration times. Now the separation coefficients can be calculated with the predicted migration times. In

this way one circumvents the direct modelling of migration times and separation coefficients. As for the peak heights, α_{\min} was selected for the whole experimental domain. The transformation of H_{\min} and α_{\min} was performed according to Eq. (1), while that of the analysis time was performed according to 'reversed' one-sided transformation. The following limits were selected for α_{\min} : $Y^- = 1.02$ (no separation) and $Y^+ = 1.065$ (desired separation); H_{\min} : $Y^- = 0.35 \cdot 10^{-2}$ V (lowest tolerable height) and $Y^+ = 0.60 \cdot 10^{-2}$ V (desired height); and t_m : $Y^- = 2.50$ min (no peaks are expected below 2.5 min, because of the mobilities of the RE) and $Y^+ = 4.00$ min (maximum analysis time below which a separation is possible normally in CE). In the transformation of the criteria α_{\min} and H_{\min} , the factor r is equal to 1.

For the migration time r is equal to 0.3, meaning that the d -scale for the analysis time between 2.5 (Y^-) and 4 min (Y^+) does not decrease linearly, but gradually decreases from 1 to 0. This gives a high quality value (Fig. 1b) to electropherograms with analysis times smaller than 4 min. Normally, as CE is considered to be a fast technique, analysis time is not expected to give problems. Still, one favours a short analysis over a longer one, as this is not only efficient in terms of time but also in costs.

The response surfaces of the scaled criteria are shown in Fig. 4a, Fig. 4b and Fig. 4c for H_{\min} , analysis time, and α_{\min} , respectively. As can be seen, H_{\min} (Fig. 4a) is optimal at conditions where the separation α_{\min} (Fig. 4c) is minimal, but still acceptable. The response surface of the overall quality (D)

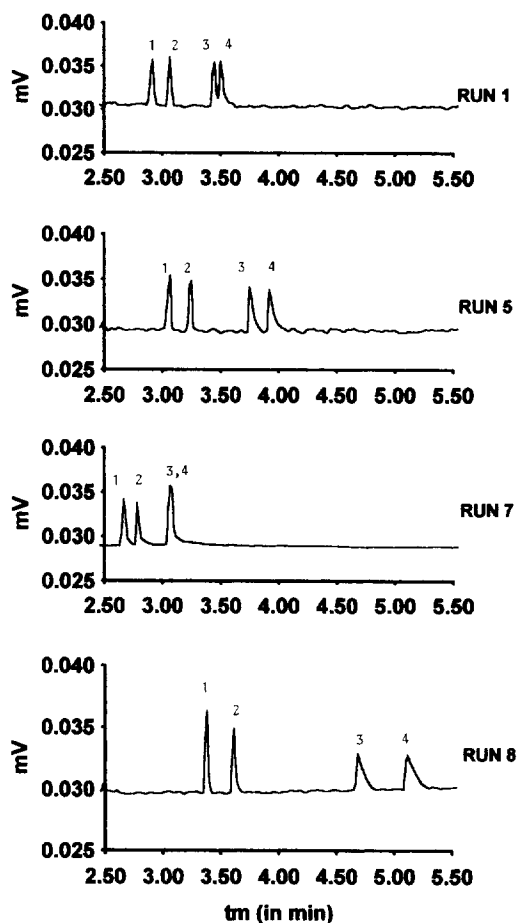


Fig. 2. The electropherograms obtained for runs 1, 5, 7 and 8 of the central composite design (Table 1). A 10 mg l^{-1} mixture of the rare earths was injected for 20 s by electromigration at 30 kV, the running voltage was 30 kV ($\pm 20 \text{ }\mu\text{A}$). Peak 1=La, peak 2=Ce, peak 3=Gd and peak 4=Tb.

is presented in Fig. 4d. The optimum conditions can now be selected. A D -value between 0.50 and 0.66 is obtained in the region pH 3.7–4.2 and [HIBA]=1.75–3.75 mM. The optimum conditions which were selected before [6] are within this region. In fact, for pH 4.1 and [HIBA]=2 mM, a D value of 0.64 is obtained, which indicates acceptable separation conditions. This resulted in the electropherogram ($D=0.67$) obtained in Fig. 3. Table 1 also shows the D values obtained for each run separately.

According to the theory [13], the outcome of this procedure only meets the specifications when $D \geq 0.37$. As can be observed, only runs 1, 2 and 5 result

in a value ≥ 0.37 , indicating acceptable conditions. This can also be seen in Fig. 2, where runs 1 and 5 have good separation within an analysis time of below 4 min and also have an acceptable peak height. Runs 7 and 8 violate the criterion of the separation or the analysis time. According to this procedure, run 1 (Fig. 2) globally approximates the overall desired requirements of the three criteria, and therefore also has the highest quality value.

3.2. Separation described by the resolution

The separation coefficient can be used as an optimization criterion only in limited cases, as it does not consider peak width. Therefore, an approach was sought for the use of a resolution-based optimization criterion such as the minimum resolution. Calculation of this criterion requires values of migration times and peak widths. The width of the peaks is not considered to be a problem as it can be determined and modelled. This is done regularly in HPLC [17]. As the migration is determined by both the effective mobility of a compound and the electroosmotic mobility, these two characteristics were modelled and the migration times were then obtained. The resolution can then be calculated with the modelled peak widths according to Eq. (4):

$$R_s = 1.117 \frac{(t_{m_2} - t_{m_1})}{(W_{1/2}^1 - W_{1/2}^2)} \quad (4)$$

where t_{m_2} and t_{m_1} are the migration times of the second and the first peak of a peak pair, and $W_{1/2}^1$, $W_{1/2}^2$ are the peak widths at half height.

Application of this procedure resulted in the data presented in Table 2. As can be observed, the predicted resolutions are not always accurate, especially for the peaks of Gd and Tb ($R_s 3$). A further investigation revealed that the inaccurate predictions of the resolution were caused by the modelling of the peak widths, which was sometimes inaccurate, especially for the last peak pair. A reason for this problem probably lies in the asymmetric peaks that are sometimes noticed.

3.3. Resolution described in electrophoretic terms

Another possible approach is to apply the frequently used expression for resolution in CE:

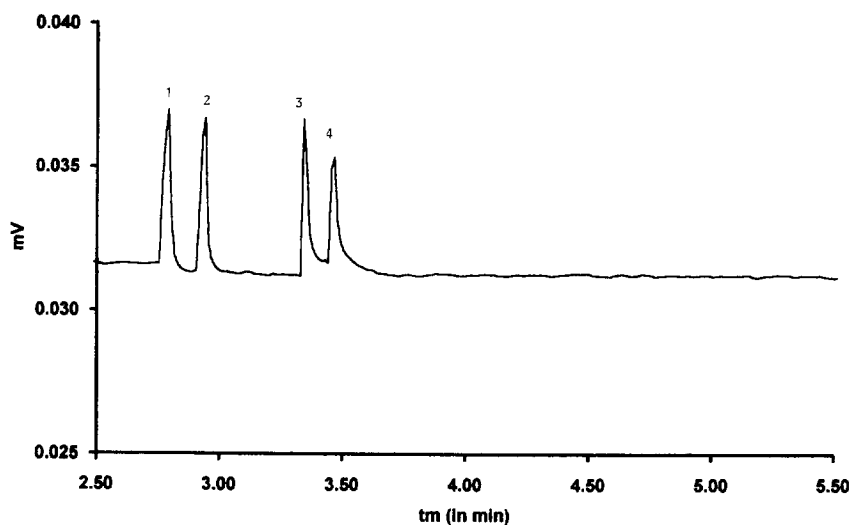


Fig. 3. Electropherogram of the REs at the predicted optimal conditions (published in [6]): 10 mM creatinine–acetate buffer, pH 4.10, and [HIBA] = 2 mM. A 10-mg l⁻¹ mixture of the rare earths was injected for 20 s by electromigration at 30 kV, the running voltage was 30 kV ($\pm 20 \mu\text{A}$.). Peak 1 = La, peak 2 = Ce, peak 3 = Gd and peak 4 = Tb.

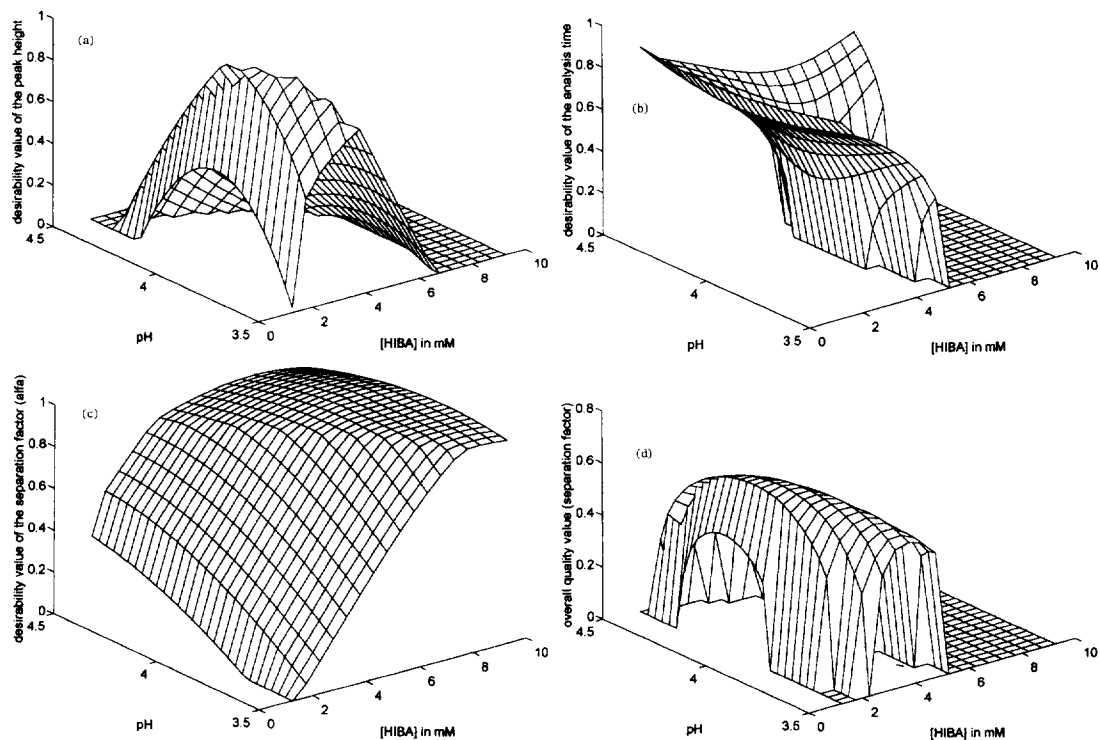


Fig. 4. Response surface of the D values for the different criteria, (a) the minimum peak height (H_{\min}), (b) the analysis time and (c) the minimum separation coefficient (α_{\min}) as a function of the pH and concentration of HIBA. The response surface of the D -values as a function of the pH and concentration of HIBA for the overall quality, when the separation coefficient is used to quantify the separation is shown in (d).

Table 2
Experimentally obtained and predicted resolutions (R_s) calculated using Eq. (4)

Run	R_s^1 exp	R_s^1 pred	R_s^2 exp	R_s^2 pred	R_s^3 exp	R_s^3 pred
	2.60	2.68	8.58	9.28	1.14	0.96
	2.58	2.65	10.99	10.41	4.25	2.76
	4.92	4.76	12.39	12.30	3.10	3.14
	5.00	4.59	16.08	15.01	3.97	3.74
	3.30	3.30	9.82	9.61	2.35	2.39
	3.16	3.14	11.72	12.18	3.30	3.79
	2.77	2.61	10.88	10.30	0.34	0.80
	5.55	6.03	14.97	15.08	3.50	3.35
	2.91	2.91	9.59	9.59	2.80	2.79

The predicted values are obtained after modelling of mobilities and peak widths at half height $W_{1/2}$.

exp, Experimental.

pred, Predicted by the model.

$$R_s = \frac{\sqrt{N_m}}{4} \cdot \frac{(\mu_2 - \mu_1)}{(\mu_m + \mu_{\text{eof}})} \quad (5)$$

where N_m is the mean of the separation efficiency obtained for two adjacent peaks, μ_2 , μ_1 and μ_m are the effective mobilities and the mean of the effective mobilities of the second and the first eluting peaks of a peak pair which is considered, respectively and μ_{eof} is the electroosmotic mobility. Eq. (5) requires prediction of the separation efficiency (N) and of the mobilities of the analytes and the neutral peak. The separation efficiency can, however, be estimated from the height and the areas of the peaks [18] according to the expression:

$$N = \frac{(2\pi \cdot t_m^2 \cdot H^2)}{Ar^2} \quad (6)$$

where Ar (in V·s) is the area of a peak. The resolution is calculated by first modelling the mobilities of the analytes and that of the EOF and also the peak areas and peak heights. Schoenmakers et al. [19] already applied a similar kind of approach in HPLC optimization studies. Eq. (6) is based on Gaussian peaks, but although this is not always true in CE, probably due to the high efficiency of the technique, the calculations still yielded acceptable results. This procedure was followed to predict the resolution of each neighbouring peak pair over the whole experimental domain. The predicted values of

Table 3
Experimentally obtained and predicted resolutions (R_s) calculated with Eq. (5), using peak areas and peak heights for the calculation of the separation efficiency

Run	R_s^1 exp	R_s^1 pred	R_s^2 exp	R_s^2 pred	R_s^3 exp	R_s^3 pred
1	5.22	5.17	14.00	13.97	1.50	1.71
2	5.83	5.91	17.61	18.28	4.68	4.09
3	7.86	7.70	22.08	21.55	3.81	3.93
4	7.67	7.64	23.87	24.03	4.44	4.89
5	6.22	6.38	15.26	15.49	3.34	3.63
6	6.78	6.80	20.62	19.97	4.99	4.94
7	5.46	5.23	16.83	15.59	0.46	0.45
8	8.05	8.14	26.28	26.70	4.91	4.38
9	8.60	8.60	24.95	24.97	6.48	6.47

The predicted values of the resolution are obtained after modelling of mobilities, areas and peak heights.

exp: Experimental.

pred: Predicted by the model.

the resolutions were compared with the experimentally obtained ones. The results are shown in Table 3. As can be seen, this approach appears to result in more accurate results than the one based on modelling of the peak widths. Therefore, it is used for the calculation of minimum resolutions which are then transformed to the D -scale using the Derringer's transformation functions. For quantitative purposes, it is desirable to achieve a resolution of at least 1.5, since this corresponds to a baseline separation of two Gaussian peaks of equal size [20]. However, in practice, a resolution of 1.25 for a peak pair with different possible peak-height ratios, is considered sufficient for an adequate separation. A separation beyond resolution 3.0 is superfluous. Based on this information, a value of 1.25 and 3.0 were selected as the lowest (Y^-) and highest (Y^+) level of the D -scale for the $R_{s_{\text{min}}}$, respectively. The obtained response surface for $R_{s_{\text{min}}}$ is presented in Fig. 5a. Considering all the D -scaled response surfaces of the peak height, analysis time and the minimum resolution, the overall quality (D) response surface can be calculated and is presented in Fig. 5b. This figure very much resembles the overall quality response surface calculated with the separation coefficient (Fig. 4d). The optimum separation conditions can now once again be selected. The optimum is situated in the same region as observed with the calculations of the separation coefficient. In the region with the limits

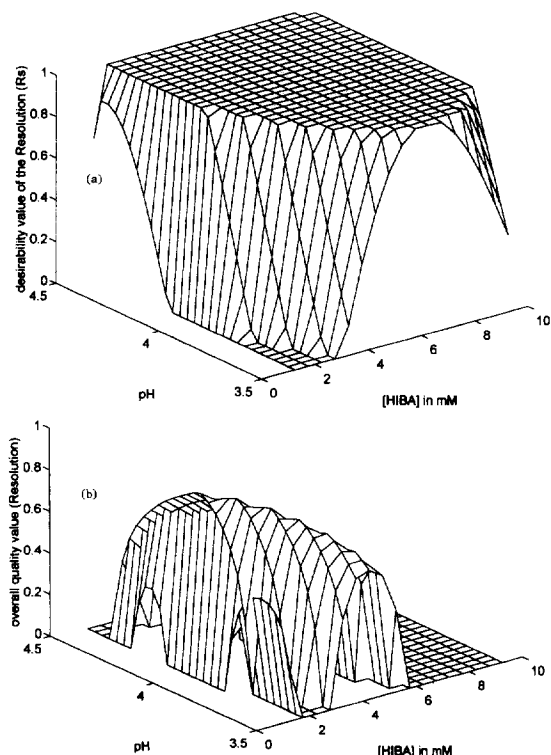


Fig. 5. Response surfaces of the D values for (a) the minimum resolution ($R_{s, \min}$) and (b) the overall quality D values when resolution is used to quantify the separation, as a function of the pH and concentration of HIBA.

for pH 3.85–4.2 and [HIBA] = 1.75–3.75 mM, a D value is obtained between 0.65 and 0.85. For the selected optimum condition in [6], a D value of 0.77 is predicted. The D value calculated for the separation in Fig. 3 resulted in a value of 0.83 (Table 1), indicating acceptable optimum separation conditions.

4. Conclusions

The introduction of Derringer's desirability function to the optimization of more than two optimization goals in CE, resulted in the selection of acceptable experimental conditions for optimal separation, sensitivity and analysis time. The selection was based on the response surface of the overall quality function, which is constructed starting from the response surfaces of each optimization criterion.

A resolution-based optimization approach is pro-

posed. The resolution is calculated after modelling of electrophoretic and electroosmotic mobilities, peak heights and areas. This procedure not only circumvents the direct modelling of migration times, but also the modelling of peak widths. The widths of the peaks are sometimes not only difficult to model, but are time-consuming to determine as well. As peak heights and areas are generally acquired by most of the recent data workstations and are printed out on custom reports, the proposed procedure is interesting from an automatization point of view.

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