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Statistical approach to model discrimination for the radical copolymerization of methyl methacrylate and styrene from *a posteriori* data of composition

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Abstract: The multiresponse maximum likelihood (MML) method and statistical tests – the *F*-test, the *t*-Student test, and the sign changes test – were compared with respect to their power of discrimination between two kinetic models, the terminal and the penultimate model, for the radical copolymerization of the styrene - methyl methacrylate monomer system in bulk. The results indicate that the investigated statistical methods (except the sign changes test) are suitable for kinetic-model discrimination when they are applied to *a posteriori* data of copolymer composition. Although the MML method was shown to be more demanding in the error estimation of experimental data, it can serve as a proper tool for kinetic-model discrimination in copolymerization, provided the correct estimates of the ratio between errors in measured copolymer composition data are known. The main steps of the calculating scheme are given.

Introduction

Since the fundamental work of Fukuda et al. [1] it is well known that the bulk copolymerization of styrene (S) and methyl methacrylate (M) conforms to the terminal model [2] with respect to composition and copolymer microstructure (triad/pentad fraction). At the same time, it entirely fails with respect to the absolute value of the propagation rate constant. This failure was ascribed to the penultimate unit of the growing radical that affects the stability of the propagating radical and thus its reactivity. To justify the successful use of the simple, and for many monomer systems enough conclusive, terminal model for composition data, the restricted (implicit) penultimate model of copolymerization was proposed [3]. A more complex model, assuming the penultimate unit effect on both, copolymer composition and overall propagation rate, introduced many years ago [4], is now called the explicit penultimate model.

The classical attempts to discriminate between implicit (terminal) and explicit penultimate kinetic models consist in testing copolymerization models against experimental data of feed and copolymer composition. These models incorporate two or four reactivity ratios treated as adjustable parameters. These attempts are often considered to be not enough conclusive, if they are conducted without thorough experimental data analysis, as described below. The reported reason is that monomer reactivity ratios are adjustable parameters in the model-fitting procedures for copolymerization experiments instead to be measured independently [5].

The methods discussed in this paper use measured copolymerization data resulting from an accomplished experiment, called by us *a posteriori* data. We use this term to distinguish these data from those of discrimination methods discussed by Burke et al. [6]. In their approach copolymer composition data come also from experiment, however, they result from an experiment designed according to experiment planning methods (e.g., a discriminative experiment planned with the aid of the D-optimization method).

As it was mentioned above the styrene - methyl methacrylate monomer system is known to be well described with the terminal model with respect to the copolymer composition data. Taking advantage of this fact we tested several statistical methods on their power in kinetic-model discrimination using *a posteriori* experimental data taken from literature. In other words our goal is not to study weather the styrene - methyl methacrylate copolymer composition data can be better or not described with the terminal (implicit) or explicit penultimate model (that is well known) but to compare the discriminative power of several selected statistical methods. Furthermore, we draw conclusions on the general reliability of kinetic-model discrimination in radical copolymerization according to studied statistical methods from the quality of error estimates of experimentally measured copolymer composition data.

Experimental part

For the reasons outlined above the system styrene - methyl methacrylate has been selected. Experimental data sets on radical copolymerization in bulk were those published by Fukuda et al. [1] and Maxwell et al. [7]. The first data set has already been proven to be accurate [8-9], with a sufficient number of experimental points and reliable estimates of error. The data of Maxwell have a small number of points and, as it will be shown below, the error estimates are probably inaccurate.

In this work four different statistical measures have been adopted for evaluation of the above data (and error estimates) and copolymerization model discrimination. These methods are: the multiresponse maximum likelihood (MML) method, Fischer's *F*-test, the *t*-Student test and the sign changes test. Above methods are of different statistical power. From a theoretical point of view the most conclusive should be the MML method provided the error estimates are correct and follow the normal distribution. The assumed normal distribution of errors does not allow for comparison with the error-in-variables model (EVM) method [10-11], which is widely used in literature. The reason is that the EVM method makes use of a covariance matrix of errors with built-in correlation between errors. The smallest discriminative power has the sign changes test. In the following we give essentials for the use of all above methods.

The use of the MML method applied to copolymer composition data analysis and model discrimination was described previously [12]. The MML method applied to copolymerization data reduction, for correct estimates of errors and a perfect model, gives the result that the most probable state is reached when the objective function, S, given by

$$S = \sum_{i=1}^{n} \left\{ \left(\frac{F_i^m - F_i}{\sigma(F_i)} \right)^2 + \left(\frac{f_i^m - f_i}{\sigma(f_i)} \right)^2 \right\} / (2n - p) \tag{1}$$

and that the relative standard deviations of both variables should be, within some probability range, in the vicinity of 1. An acceptable set-off depends on the probability level assumed. In Eq. (1), $\sigma(f_i)$ and $\sigma(F_i)$ are error estimates in both variables, f_i the monomer feed, and F_i the copolymer compositions, respectively. Superscript m stands for experimental (measured) values whereas variables without superscript are estimated; n stands for the number of experimental points and p for the number of adjustable parameters. In the MML method, errors in all variables are estimated together with the parameters of the model used. Therefore, the number of adjustable parameters p in Eq. (1) is equal to the number of experimental points plus the number of adjustable parameters in the copolymerization model applied. The relative standard deviation of variables is given by Eq. (2),

$$rel.\sigma(y) = \sqrt{\sum_{i=1}^{n} \left[\left(y_i^m - y_i\right)/\sigma(y_i)\right]^2 / (n-p)} \qquad (2)$$

where y stands for either f or F. The remaining symbols are the same as in Eq. (1), but p in this case is equal to the number of adjustable parameters in the copolymerization model. Standard deviation $\sigma(F)$, reported in Tab. 1, for data fitted with the least squares method, LSQ, was calculated with the formula without $\sigma(F_i)$.

The F-test for copolymerization model discrimination was introduced first by Hill et al. [13]. Comprehensive background for the use of the F-test was given by Burke et al. [6]. The same method was later applied to the monomer system in solution [14]. Using the terminal and penultimate models for experimental data reduction we have two independent distributions of differences between experimental and calculated values of variables. Assuming that differences obtained for both models are normally distributed populations, we can use the F-test for testing the null hypothesis that the 2nd model with smaller standard deviation, $\sigma_2(F)$, differs from the other one insignificantly. To this end the test variable is calculated according Eq. (3), where corresponding models have been denoted as 1 and 2.

$$F = \frac{\sigma_1^2}{\sigma_2^2} \quad \text{with } \sigma_1 > \sigma_2 \qquad (3)$$

The assumed null hypothesis is fulfilled with an error probability $1 - \alpha$ if the calculated test value F is less than the critical value $F_c(\alpha, n - p_1, n - p_2)$, obtained using the F-distribution for probability level α and $n - p_1$ and $n - p_2$ degrees of freedom.

The terminal and penultimate models have two and four reactivity ratios, respectively, regarded as adjustable parameters fitted to the experimental copolymerization data. We already mentioned shortly elsewhere [15] that, if the least square technique is used for solving the fitting job, as a by-product the so-called variance-covariance matrix could be obtained (for the adjustable parameters denoted for short as r)

$$Cov(r) = \sigma^2(F)\,(J^T J)^{-1} \qquad (4)$$

where J stands for the Jacobian (matrix of partial derivatives of dependent variables with respect to the adjustable parameters at current values of adjustable parameters), superscript 'T' indicates the transposed matrix of J, and superscript '-1' indicates the inverse matrix. The diagonal elements of the $Cov(r)$ matrix consist of the variance values of corresponding reactivity ratios, i.e., the square roots of these elements are equal to the estimates of standard errors of reactivity ratios. With these estimates, the t-Student test can be used to test the null hypothesis that a particular calculated reactivity ratio differs significantly from zero. To this end the test value t must be calculated

$$t = \frac{|r_i|}{\sigma(r_i)} \tag{5}$$

where r_i stands for the tested adjustable parameter (reactivity ratio for the particular model) and $\sigma(r_i)$ for the estimated standard deviation of this parameter.

The assumed null hypothesis is fulfilled with an error probability $1 - \alpha$ if the calculated test value t is greater than the critical value $t_c(\alpha, n - p)$, obtained using the t-distribution for probability level α and $n - p$ degrees of freedom. If the null hypothesis is fulfilled one speaks of significance of the parameter.

Information contained in the covariance matrix may be used in another way for model discrimination. This is connected with the use of correlation coefficients between parameters of the given model. The correlation coefficient ρ_{ij} for the pair of parameters r_i and r_j is defined as

$$\rho_{ij} = Cov(i,j)/\sqrt{Cov(i,i)Cov(j,j)} \tag{6}$$

The value of this coefficient may range from 0 to ±1. The value of correlation coefficients in the vicinity of ±1 indicates that the relevant parameters r_i and r_j are, probably, linearly dependent and one of them should be removed from consideration.

Deviations between experimental and calculated values represent statistically distributed variables. Assuming the normal distribution, the sign changes test can be applied to these data to test for systematic trends. If two neighbouring deviations have opposite sign, then one speaks of a sign change. A measure of the randomness of deviations is the number of sign changes. The expected number should be within the range $n/2 \pm (n/2)^{0.5}$ at the 68% confidence level. As can be seen from results reported in Tab. 1, the sign changes test has no discriminative power in the case of the investigated data.

Finally, it should be emphasized that to perform any of the statistical tests, e.g., the F-test or the t-Student test, data should be processed using suitable methods of fitting the adjustable parameters in a given model to the experimental data. To this end the MML method can be recommended but also other methods can be used, in particular the least squares method, LSQ, or the weighted LSQ method.

Results and discussion

Tab. 1 contains results of experimental data processing. The table reports calculated reactivity ratios for the terminal and/or penultimate model, r_i or r_{ij}, their standard errors, $\sigma(r_i)$ or $\sigma(r_{ij})$, the standard deviation of the objective function, $\sigma(S)$, the relative standard deviations of monomer feed and polymer compositions, $rel.\,\sigma(f)$ and $rel.\,\sigma(F)$, the relative standard deviation of the objective function, $rel.\,\sigma(S)$, and the values of the sign changes test. Data were calculated using the MML method (terminal model) and the LSQ method (both, terminal and penultimate model). The MML method in principle can be used also for the penultimate (or any other suitable) model. However, its use has been excluded due to the non-uniqueness in determination of the penultimate model parameters for the styrene - methyl methacrylate copolymerization system in bulk discussed in one of our previous papers [9].

In the MML calculation, for data by Fukuda et al. [1], we used error estimations equal to 0.015 for both variables, f_i and F_i. These values are equal to original estimates given by the authors (1.5%). Maxwell et al. [7] estimated for their composition data set the random error (precision) in the experimentally observed dependent and independent variables as 5% for copolymer composition (calculated from NMR peak areas) and 1% for the fraction in the feed. As argued above, for correct estimates of errors and a perfect model, the calculated values of standard deviation should be in the vicinity of 1 for MML.

Tab. 1. Validation of the kinetic model. This table reports calculated reactivity ratios for the terminal or terminal and penultimate model, r_i or r_{ij}, their standard errors, $\sigma(r_i)$ or $\sigma(r_{ij})$, the standard deviation of the objective function, $\sigma(S)$, the relative standard deviations of monomer feed and polymer compositions, *rel.* $\sigma(f)$ and *rel.* $\sigma(F)$, the relative standard deviation of the objective function *rel.* $\sigma(S)$, and the values of the sign changes test (SCT)

	r_i	$\sigma(r_i)$	$\sigma(S)$	*rel.* $\sigma(f)$	*rel.* $\sigma(F)$	*rel.* $\sigma(S)$
			SCT	SCT	SCT	
			MML method			
Fukuda et al. [1]						
r_S	0.5460	0.0090		0.51016	0.57103	0.54145
r_M	0.4822	0.0089		9 [a]	9 [a]	
Maxwell et al. [7]						
r_S	0.4638	0.0140		0.04458	0.24495	0.17605
r_M	0.4083	0.0080		5 [b]	5 [b]	
			LSQ method			
Fukuda et al. [1]						
r_S	0.5500	0.0180	0.01113			
r_M	0.4816	0.0171	11 [a]			
r_{SS}	0.6599	0.0850	0.01089			
r_{MM}	0.4862	0.0550	11 [a]			
r_{MS}	0.3708	0.1022				
r_{SM}	0.3887	0.1046				
Maxwell et al. [7]						
r_S	0.4641	0.0282	0.00895			
r_M	0.4045	0.0160	5 [b]			
r_{SS}	0.9614	0.6438	0.00937			
r_{MM}	0.4542	0.0657	4 [a]			
r_{MS}	0.2004	0.1425				
r_{SM}	0.2664	0.0881				

[a] Statistically correct values should be within (6,12).

[b] Statistically correct values should be within (2,6).

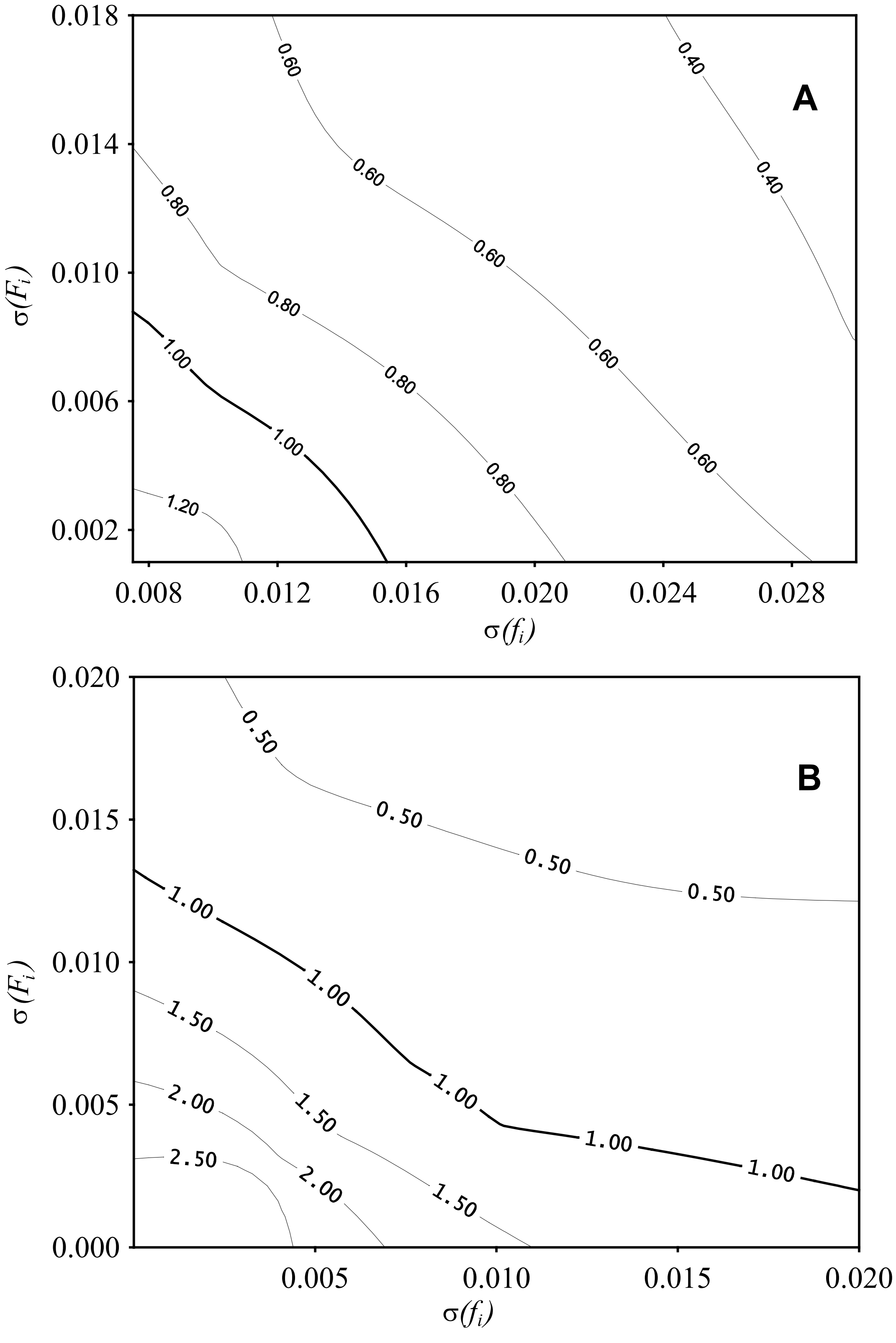

Fig. 1. Planar ($\sigma(f_i)$, $\sigma(F_i)$) projection of the 3-dimensional space ($\sigma(f_i)$, $\sigma(F_i)$, *rel*. $\sigma(S)$). Lines correspond to the same values of *rel*. $\sigma(S)$. Bold lines indicate curves for *rel*. $\sigma(S) = 1$. Scanning results represented for: (A) Data by Fukuda et al. [1], scanned in the ranges $\sigma(f_i)$ <0.008, 0.028>, $\sigma(F_i)$ <0.001, 0.018>. (B) Data by Maxwell et al. [7], scanned in the ranges $\sigma(f_i)$ <0.001, 0.020>, $\sigma(F_i)$ <0.001, 0.020>. All scanning calculations were performed with steps equal to 0.001

From Tab. 1 it follows that the standard deviation (Eq. (1)) as well as the relative standard deviations of monomer feed and copolymer compositions for the MML calculations are far from 1 and outside of the confidence interval. At the 95% confidence level, the corresponding values are equal to 0.65 and 1.35, and 0.51 and 1.49, for data by Fukuda et al. and Maxwell et al., respectively. This indicates either the non-applicability of the terminal model to the data, or incorrect estimation of the errors. As it was stated above it is generally accepted that the bulk copolymerization of the S/M monomer system conforms to the terminal model.

For data by Fukuda et al., the standard deviation of the objective function, $\sigma(S)$, the relative standard deviations of monomer feed and polymer compositions, *rel.* $\sigma(f)$ and *rel.* $\sigma(F)$, are nearly equal and about two times lower than 1. This suggests that the error estimates are about twice overestimated. For data by Maxwell et al. standard deviation estimates are also far from unity but not of the same magnitude. Therefore it may be suspected that Maxwell's estimates of the error were incorrect.

We tried to check both above statements. Provided the terminal model of copolymerization is the adequate one, we can try to find the 'right' error estimates by scanning the 3-dimensional space ($\sigma(f_i)$, $\sigma(F_i)$, $\sigma(S)$) and looking for $\sigma(f_i)$ and $\sigma(F_i)$ values corresponding to $\sigma(S) = 1$ (as well as $\sigma(f) = 1$ and $\sigma(F) = 1$). Results are presented in Fig. 1.

For data by Fukuda et al., the optimal values $\sigma(f_i) = 0.0082$ and $\sigma(F_i) = 0.0078$ result in the following estimates: $\sigma(S) = 1.02$, $\sigma(f) = 0.98$, and $\sigma(F) = 1.05$. These values satisfy the demands for application of MML. It should be noted that the ratio of estimates of errors (0.0082/0.0078) is similar to the estimation given by the authors (0.015/0.015).

For data by Maxwell et al., the optimal values $\sigma(f_i) = 0.008$ and $\sigma(F_i) = 0.006$ result in the following estimates: $\sigma(S) = 1.016$, $\sigma(f) = 1.009$, and $\sigma(F) = 1.023$. These values also satisfy the demands for application of MML. However, contrary to data by Fukuda et al., the ratio of estimates of errors (0.008/0.006) is quite different from the ratio calculated from values reported by the authors (0.01/0.05).

Calculations with the MML support the applicability of the terminal model for both sets of data but indicate the improper error estimation for data by Maxwell et al. It must be stressed that from a statistical point of view the most important is the proper estimation of the relation between relevant errors in variables instead of their absolute values.

For the reasons mentioned above the results obtained from the LSQ method were used for the *F*-test and the *t*-Student test.

For data by Fukuda et al. ($n = 18$) the standard deviation corresponding to the penultimate model ($\sigma(F) = 0.01089$) is less than the standard deviation for the terminal model ($\sigma(F) = 0.01113$). To test the null hypothesis that the penultimate model represents experimental data significantly better, the F value has to be calculated: $(0.01113/0.01089)^2 = 1.0446$. The corresponding F_c value for $\alpha = 0.05$ (95% probability level) and 16 and 14 degrees of freedom is equal to 2.46. The calculated test value is less than the critical one. This means that the null hypothesis should be rejected. Taking into account that the penultimate model claims four adjustable parameters, it follows that a model using less parameters, i.e., the terminal one, has to be adopted. For data by Maxwell et al. ($n = 8$) the standard deviation corresponding to the penultimate model ($\sigma(F) = 0.00937$) is higher than the standard deviation for the terminal model ($\sigma(F) = 0.00855$). Thus, application of the *F*-test is only formal; the model with smaller standard error and number of parameters should be preferred. The *F*-test applied to these data also indicates that both models differ insignificantly ($(0.00937/0.00855)^2 = 1.201 < 6.16$). Thus, the *F*-test shows that the terminal model should be preferred in both cases.

The parameters for both kinetic models of copolymerization reported in Tab. 1 were tested for their significance, i.e., if the parameters are significantly different from zero, from a statistical point of view. This is fulfilled if the calculated t value is greater than the critical value, $t_c(\alpha, n - p)$. For data by Fukuda et al. it was found that all parameters are significant at $\alpha = 0.05$ (95% probability level) whereas for data of Maxwell et al. two insignificant parameters, r_{SS} and r_{MS}, were evaluated. For the r_{SS} the calculated test value is $t = 0.9614/0.6438 = 1.493$ whereas $t_c(\alpha = 0.05, n - p = 4) = 2.776$. Since $t < t_c$, r_{SS} insignificantly differs from zero. For r_{MS} we have $t = 0.2004/0.1425 = 1.406 < 2.776$ ($\alpha = 0.05$, $n - p = 4$). This conclusion is supported by analysis of the correlation coefficients. For example, $\rho(r_{SS}, r_{MS}) = -0.9834$.

It can be concluded that for data by Fukuda et al. the *t*-test has no discriminative power whereas for the second data set the test indicates insignificance of two parameters. This can be understood as encouragement to use the terminal model.

Conclusions

The two data sets selected from literature are of different reliability, i.e., accuracy, number and distribution of experimental points and reliability of error estimates reported by the authors. Nevertheless, all the statistical tests (except the sign changes test) applied to these *a posteriori* experimental data sets unambiguously indicate that the terminal model is sufficient for description of the data within estimated experimental error. The sign changes test, in this particular case, has no discriminative power. It is also clear that the researchers should take care for more thorough analysis of the uncertainties of provided data. This demand is particularly important when more sophisticated statistical techniques have to be used. Therefore, for successful use of the MML method, the correct estimates of the errors in all measured variables must be available. However, as already concluded [12], from a statistical point of view, the most important is the proper estimation of the relation between relevant errors in variables instead of their absolute values. This agrees with the opinion of van Herk et al. [16] on the importance of the error structure in determining the suitability of nonlinear least squares fitting procedures to the modelling of radical copolymerization.

Finally, it should be noted that combining different ‘statistical tools’ substantially increases the reliability of model discrimination.

Appendix

Theoretical background on the use of the MML method applied to copolymer composition data analysis was given previously [10]. The problem consists in the minimization of the objective function S given by Eq. (1). The objective function has to be minimized with respect to both, monomer feed, f_i, and adjustable parameters, r_i, of the corresponding (terminal or penultimate) model.

To solve the nonlinear problem, we assume the monomer feed, f_i, and reactivity ratios, r_j, to be independent variables, and the copolymer composition, F_i, be the dependent variable. The depending variable, F_i, has to be linearized with respect to monomer feed and reactivity ratios (cf. Eq. (6) in ref. [10]). A set of $2n$ equations with $n + 2$ unknowns is obtained. The unknowns are the differences between the new and next recent values of independent variables. This set of equations has to be solved. The iterative procedure is sketched below.

The system of equations can be comprehensively expressed as $J \cdot \vartheta = b$, where J is the Jacobian of partial derivatives of the linearized objective function with respect to independent variables ϑ^T; $\vartheta^T = (\Delta f_i, \Delta r_j)$ denotes the transposed vector of unknowns, b ('the right hand vector') stands for a vector of weighted differences between experimental and calculated copolymer composition and monomer feed.

In the following the main steps of the calculating scheme are outlined:

1) Enter input data: experimental monomer feed, f_i, and copolymer composition, F_i, together with corresponding estimates of errors, $\sigma(f_i)$ and $\sigma(F_i)$.

2) Enter guess values for the reactivity ratios, r_j. For the terminal model, $j = s, m$; for the penultimate model, $j = ss, mm, ms, sm$.

3) Enter values for stop-criteria: *eps*, *max_iter*, *max_dr*. Set loop control variable *iter* = 0. Set $S_{\text{old}} = 10^6$.

4) Set guess values for the calculated monomer feed, f_i^{c}, and the calculated copolymer composition, F_i^{c}: for $i = 1...n$ $f_i^{\text{c}} = f_i$ $F_i^{\text{c}} = F_i$

5) Calculate the Jacobian and the 'right-hand vector' b. The Jacobian is a $2n$ x $(2n + p)$ matrix, where n is the number of experimental points and p the number of adjustable parameters [$p = n$ + (number of reactivity ratio parameters)].

a) for $i = 1...n$; $j = 1...n$

$$J_{ij} = \frac{1}{\sigma(F_i)} \frac{\partial F_i}{\partial f_i}; \quad i = j \qquad\qquad J_{ij} = 0; \quad i \neq j$$

b) for $i = 1...n$; $j = n + 1 ... n + p$

$$J_{ij} = \frac{1}{\sigma(F_i)} \frac{\partial F_i}{\partial r_{j-n}}$$

c) for $i = n + 1 ... 2n$; $j = 1...n$

$$J_{ij} = \frac{1}{\sigma(f_i)}; \quad i - n = j \qquad\qquad J_{ij} = 0; \quad i - n \neq j$$

d) for $i = n + 1 … 2n$; $j = n + 1 … n + p$

$$J_{ij} = \frac{1}{\sigma(f_i)} \frac{\partial f_i}{\partial r_{j-n}}$$

The elements of the right-hand vector are:

for $i = 1...n$ $$b_i = \frac{F_i - F_i^c}{\sigma(F_i)}$$

for $i = n + 1 ... 2n$ $$b_i = \frac{f_{i-n} - f_{i-n}^c}{\sigma(f_{i-n})}$$

6) Solve for the unknown ϑ. To this end, the orthogonalization method can be recommended, e.g., that of Gram and Schmidt.

7) Add the calculated differences of the independent variables to the corresponding most recent values:

for $i = 1...n$ $\quad f_i^c = f_i^c + \Delta f_i$

for $j = 1...p$ $\quad r_j = r_j + \Delta r_j$

8) Check for convergence. The stop criteria should be used:

a) if $S_{\text{old}} - S < eps$ then convergence achieved; go to (9)

b) if $iter > max\_iter$ then (NO CONVERGENCE); exit procedure

c) if $max(\Delta r_j) < max\_dr$ then go to (9)

$iter = iter + 1$; $S_{\text{old}} = S$ then go to (5)

9) Convergence achieved.

NO CONVERGENCE: Two cases should be considered:

1. The initial estimates for reactivity ratios are too far from reasonable values. However, according to our experience, for the terminal model convergence is achieved using any arbitrary pair of estimates.

2. For the penultimate model, additionally non-uniqueness in the determination of the reactivity ratio parameters should be taken into account as discussed previously in ref. [9].

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