Journal of Macromolecular Science, Part A
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597274

Analysis of the Linear Methods for Determining Copolymerization Reactivity Ratios. V. Planning of Experiments

F. Tüdös[a]; T. Kelenm[b]
[a] Department of Chemical Technology, Eötvös Lòrànd University, Budapest, Hungary [b] Central Research Institute for Chemistry of the Hungarian Academy of Sciences, Budapest, Hungary

To cite this Article Tüdös, F. and Kelenm, T.(1981) 'Analysis of the Linear Methods for Determining Copolymerization Reactivity Ratios. V. Planning of Experiments', Journal of Macromolecular Science, Part A, 16: 7, 1283 — 1297
To link to this Article: DOI: 10.1080/00222338108063235
URL: http://dx.doi.org/10.1080/00222338108063235

J. MACROMOL. SCI.–CHEM., A16(7), pp. 1283-1297 (1981)

Analysis of the Linear Methods for Determining Copolymerization Reactivity Ratios. V. Planning of Experiments

F. TÜDŐS

Department of Chemical Technology
Eötvös Loránd University
H-1088 Budapest, Hungary;
Central Research Institute for Chemistry of the Hungarian Academy of Sciences
H-1525 Budapest, Hungary

T. KELEN

Central Research Institute for Chemistry of the Hungarian Academy of Sciences
H-1525 Budapest, Hungary

ABSTRACT

A method for planning of experiments has been elaborated which makes it possible to decide the adequacy of the two-parameter model of binary copolymerization. At the same time, the procedure provides equal and reasonably low relative errors of r_1 and r_2. In the approximate knowledge of the parameters and of the value of analytical error, the procedure furnishes the number of measurements and the monomer feed values necessary for the desired accuracy. It was obtained as a "rule of thumb" that, in spite of the erroneous practice, the points should be arranged uniformly within the range of copolymer composition instead of monomer composition. For the objective performance

of retrospective evaluations, an "efficiency factor" has been introduced.

INTRODUCTION

In the previous parts [1, 2] of our series of publications, a procedure was given for the determination of copolymerization constants. Owing to its linear and graphic feature, this method is especially advantageous for deciding the adequacy of the two-parameter copolymerization model. The method for the calculation of confidence intervals of copolymerization constants determined by the above procedure was also given recently [3]. Reexamination of the data of numerous carbocationic systems published in the literature was carried out [4, 5]. It was found that from the same number of experimental data and with the same measuring accuracy, diversely precise copolymerization parameters can be determined, depending on the monomer compositions at which the copolymer compositions are examined. Similarly, the proper choice of monomer-mixture compositions are necessary for the decision of adequacy on the basis of experimental data.

Although Behnken [6] and Tidwell and Mortimer [7, 8] stressed the importance of planning the experiments, the two-point design proposed by them cannot be considered as a satisfactory solution, and that is probably why it has not come into general use in the practice. Since the present practice of experimentists is far from satisfactory, we wish to contribute with this paper to the optimal planning of experiments.

AIM OF PLANNING

In the examination of a binary copolymerization system which is not yet controlled experimentally, one can never be sure whether the behavior of the system can adequately be described by the two-parameter (i.e., the simplest copolymerization) model. The irregular behavior can have many reasons, some of them (e.g., penultimate effect, complex or salt formation) have been detailed in Ref. 1. Therefore, in a careful copolymerization examination, it must be required that the experimental data (1) be suitable for the decision of adequacies of the experimental model, and (2) make it possible to determine the values of r_1 and r_2 with equal and acceptably low error.

The planning of experiments is necessary for the fulfillment of these conditions and, moreover, with a minimal number of experiments.

The two-point design and the sequential two-point design procedure [6, 7] reduce the examination of a composition region to two points, thus excluding in advance the examination of adequacy. This is analogous to the procedure of linear estimation theory, i.e., the case in which the linearity of the problem is given per definitionen and so the question of adequacy does not even arise. Since, according to the above-mentioned facts, this is not assumed in most cases [5], the two-point design method is generally not applicable, and it should be replaced by a method which makes it possible to have a clear-cut standpoint on the question of adequacy. Naturally, some previous information is needed for the planning of experiments. The procedure to be treated further assumes the presence of approximate information regarding the values of r_1 and r_2, as well as the error of the determination of polymer composition.

THE PROPOSED METHOD OF PLANNING

Proof of Adequacy

The decision of adequacy of the two-parameter copolymerization model is based on the estimation of linearity of the K-T transformation. In the case of graphic evaluation it is highly facilitated if the n experimental points are located symmetrically and equidistantly in the figure.

Symmetrical location of the points means that

$$\xi_i = 1 - \xi_{n-i+1} \qquad (1)$$

$(1 \leqq i \leqq n)$ and equidistant location means that

$$\Delta\xi = \xi_{i+1} - \xi_i = \text{constant} \qquad (2)$$

The fulfillment of conditions (2) and (3) involves

$$\Sigma\xi_i = \Sigma(1 - \xi_i) = n/2 \qquad (3)$$

and

$$\Sigma\xi_i^2 = \Sigma(1 - \xi_i)^2 \qquad (4)$$

Requirement of Equal Error

As learned from earlier studies (see, e.g., Refs. 7 and 8), the errors of r_1 and r_2 values calculated from a given set of measurements can be highly different in the case of an improper planning of experiments. This situation is further worsened by an improper evaluation procedure [1, 6-8]; a known example for this is the linearization introduced by Fineman and Ross [9]. Reliable planning of copolymer compositions and elaboration of different theories of reactivity require, however, that the data base–the set of values of copolymerization parameters–be composed from elements of approximately equal reliability. That means that in a well-planned experiment it should be required that the r values obtained from the experimental data have approximately equal relative error:

$$\Delta r_1/r_1 = \Delta r_2/r_2 \tag{5}$$

As it was pointed out in Ref. 3, the 95% confidence intervals of the copolymerization constants can be given by the following expressions:

$$\Delta r_1 = \pm t_{0.95}(n-2)\sqrt{\frac{S^2_{\min}}{n-2}\,\frac{\Sigma(1-\xi_i)^2}{D}} \tag{6}$$

$$\Delta r_2 = \pm \alpha t_{0.95}(n-2)\sqrt{\frac{S^2_{\min}}{n-2}\,\frac{\Sigma\xi^2}{D}} \tag{7}$$

where $S^2_{\min}$ is the minimum value of the residual sum of squares:

$$S^2 = \Sigma[\eta_i - r_1\xi_i + \frac{r_2}{\alpha}(1-\xi_i)]^2 \tag{8}$$

and

$$D = \Sigma\xi_i^2 \cdot \Sigma(1-\xi_i)^2 - [\Sigma\xi_i(1-\xi_i)]^2 \tag{9}$$

and $t_{0.95}(n-2)$ is the tabulated value of the Student distribution for n data on the 0.95 probability level.

Accordingly, Expressions (6) and (7) are invariant to the reindexing of the monomers, and this means that the value of $S^2_{\min}$ changes upon reindexing: $S^2_{\min}(2, 1) \rightarrow \alpha^2 S^2_{\min}(1, 2)$.

As a result of the fulfillment of Relationships (3) and (4), the following is given from Relationships (6) and (7):

$$\Delta r_2 = \alpha \Delta r_1 \tag{10}$$

which, upon substitution into (5), gives

$$\Delta r_1/r_1 = \alpha\,\Delta r_1/r_2 \tag{11}$$

and from this, upon simplification, we obtain

$$\alpha = r_2/r_1 \tag{12}$$

Consequently, a series of experiments is well planned if α_{exp} calculated from the data equals the theoretical α value defined by Eq. (12). As is known from Ref. 1, the value of α_{exp} is

$$\alpha_{\mathrm{exp}} = \sqrt{F_{\mathrm{min}} \cdot F_{\mathrm{max}}} \tag{13}$$

Requirement of Reasonably Low Error

Despite all the experimentalist's efforts, copolymer composition data can be determined only with more or less error. Systematic errors are very difficult to recognize and they can generally not be eliminated. Therefore, we do not deal with taking systematic errors into account in the planning of experiments. From among the random errors of the analysis, we are going to deal with the case of constant absolute error ($\pm\Delta$ = const). The case of constant relative error in most aspects does not substantially differ from this.

The calculation results shown in Fig. 1 indicate that in the η-ξ diagram, above and under the theoretical line corresponding to the precise composition data (according to $+\Delta$ and $-\Delta$ deviations), an error contour is formed which has a funnel-shape widening at both ends of the figure. It can be proved that if α is chosen according to Eq. (12), the contour curves have a central symmetry to the point $\xi = 0.5$, $\eta = 0$.

According to the linear estimation theory, two parameters of a straight line can be determined with maximal accuracy if the distance between the points is maximal. Therefore it would be desirable that the extreme values of ξ (ξ_{min} and ξ_{max}) approach the $\xi = 0$ and $\xi = 1$ limits as much as possible. On the other hand, the more extreme the composition of the copolymer investigated, the higher the inaccuracy caused by the error of the analysis. In the proximity of $\xi = 0$ and $\xi = 1$ values, the error funnel widens rapidly.

A compromise to solve this dilemma is that the values of ξ_{min} and ξ_{max} are chosen so that they should remain inside the range indicated by the extremes of the contour curves. A satisfactory approach for this in the case of choice $\alpha = r_2/r_1$ is

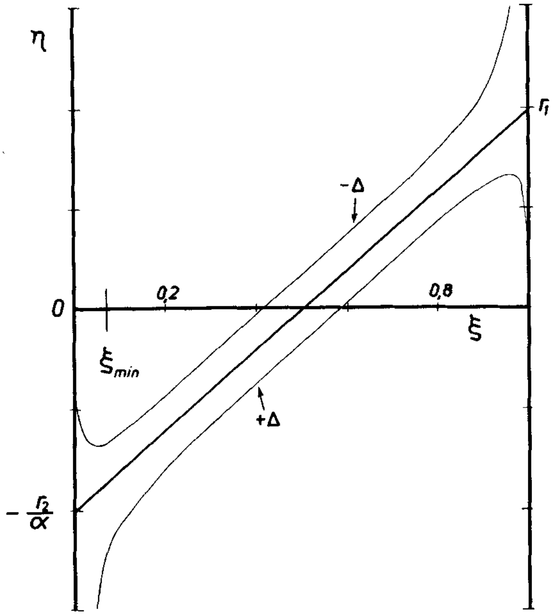

FIG. 1. KT diagram and the error contours corresponding to the analytical error $\Delta = \pm 0.03$ in the case of $r_1r_2 = 0.3$ with the choice $\alpha = r_2/r_1$. The value of ξ_{min} is also indicated.

$$\xi_{min} = 1 - \xi_{max} = \frac{\sqrt{2r_1r_2\Delta} + \Delta}{2 + 2\sqrt{2r_1r_2\Delta} + \Delta} \tag{14}$$

where Δ is the error of polymer composition expressed as mole fraction. It is remarkable that the value of ξ_{min} does not depend on the individual values of r_1 and r_2; it depends only on their product. The interval can be widened by decreasing the error of the analysis since

$$\lim_{\Delta \to 0} \xi_{min} = 0 \tag{15}$$

Number of Experiments

One can optimally experimentalize by carrying out a suitable number of experiments (i.e., not too many, not too few) for the determination of the desired accuracy of the parameters. Let us denote the latter by δ:

$$\delta = \frac{\Delta r}{r} = \sqrt{\frac{\Delta r_1}{r_1}\,\frac{\Delta r_2}{r_2}} \qquad (16)$$

Then, according to Eqs. (6) and (7), in the case of $\alpha = r_2/r_1$ and considering Eq. (4), we obtain

$$\delta^2 r_1^2 = t_{0.95}^2 (n - 2) \frac{S_{\min}^2}{n - 2}\,\frac{\Sigma\xi_i^2}{D} \qquad (17)$$

Determination of the number of measurements required for copolymerization parameters having δ accuracy can be done by Eq. (17). As can be proved by more detailed calculations, the quantities in the right-hand side of Eq. (17) depend on the number of measurements as follows:

$$\Sigma\xi_i^2 = \frac{n(n + 1)}{3(n - 1)} \left[\frac{2n - 1}{2(n + 1)} - \xi_{\min}\cdot\xi_{\max} \right] \qquad (18)$$

$$D = \frac{n^2 (n + 1)}{12(n - 1)} [1 - 4\cdot\xi_{\min}\cdot\xi_{\max}] \qquad (19)$$

and $S_{\min}^2$ can be approximated by the sum of squares of the analytical errors:

$$S_{\min}^2 \simeq 4n\Delta^2 r_1^2 \left(1 + \frac{1}{\sqrt{r_1 r_2}} \right)^2 \qquad (20)$$

With the above relationships we obtain

$$\delta^2 = t_{0.95}^2 (n - 2) \frac{16\Delta^2}{n - 2}\, \frac{\dfrac{2n - 1}{2(n + 1)} - \xi_{\min}\cdot\xi_{\max}}{1 - 4\cdot\xi_{\min}\cdot\xi_{\max}} \left(1 + \frac{1}{\sqrt{r_1 r_2}} \right)^2 \qquad (21)$$

from which the value of n can be expressed as

$$n = 2 + t_{0.95}^2 (n - 2) \frac{16\Delta^2}{\delta^2}\, \frac{\dfrac{2n - 1}{2(n + 1)} - \xi_{\min}\cdot\xi_{\max}}{1 - 4\cdot\xi_{\min}\cdot\xi_{\max}} \left(1 + \frac{1}{\sqrt{r_1 r_2}} \right)^2 \qquad (22)$$

The dependence of $t_{0.95}(n - 2)$, the Student distribution included in this expression on the n value, is usually given in tabulated form. Its limiting value is:

$$\lim_{n \to \infty} t_{0.95}(n - 2) = 1.96 \tag{23}$$

According to numerical calculations, the asymptote of the function in Eq. (22) can be approximated in the case of n ≧ 5 by the following relationship:

$$n = 3.5 + \frac{60\Delta^2}{\delta^2} \frac{1 - \xi_{min} \cdot \xi_{max}}{1 - 4\xi_{min} \cdot \xi_{max}} \left(1 + \frac{1}{\sqrt{r_1 r_2}}\right)^2 \tag{24}$$

As can be seen from Eq. (22), in the case of absolutely precise measurements ($\Delta = 0$), $n = 2$ is obtained, which is trivial. In such a case, so many measurements are really sufficient for the determination of the two parameters that it is not possible to decide the question of adequacy. In our view, five or six data measurements are necessary for a decision of adequacy, even when the measurements are accurate and even when Eq. (24) gives the lower number of measurements.

If, however, Eq. (24) gives $n > 20$, this indicates that an unreal accuracy requirement is required for the value triplet r_1, r_2, and Δ. To obtain a more realistic n value, either the accuracy requirements of δ should be reduced or the precision of the analysis should be increased, i.e., attempts should be made to decrease the value of Δ. This is a very effective method because n depends quadratically on the error of analysis.

Calculation of Monomer Composition

After having determined the number of measurements and having knowledge of ξ_{min}, we can calculate the ξ_i abscissa values of the KT plot by representing the monomer mixture compositions at which experiments should be carried out:

$$\xi_i = \xi_{min} + (i - 1)\Delta\xi \tag{25}$$

where

$$\Delta\xi = \frac{\xi_{max} - \xi_{min}}{n - 1} = \frac{1 - 2\xi_{min}}{n - 1} \tag{26}$$

The mole fractions (χ_i) of the 2 monomer in the initial monomer mixtures can be calculated by the following quadratic equation:

$$a\chi_i^2 + b\chi_i + c = 0 \tag{27}$$

with the coefficients

$$a = (r_1 + r_2 - 2r_1r_2)\xi_i - r_1(1 - r_2) \tag{28}$$

$$b = r_1(2 - r_2) - 2r_1(1 - r_2)\xi_i \tag{29}$$

$$c = -r_1(1 - \xi_i) \tag{30}$$

As an example, Table 1 shows the values of ξ_i and χ_i in the case of different r_1 and r_2 parameters ($n = 7$ and $\Delta = 0.015$).

Procedure

Based on the principles detailed above, the experiments required for the study of a binary copolymerization system can be planned as follows.

1. Collect and critically evaluate the initial informations, i.e., estimate the Δ error of the analysis as well as the approximate values of the r_1 and r_2 parameters. The latter can either be taken from the literature or some experiments should be made for their determination.
2. Based on the approximate parameters of the system, on the possibilities of analysis, and on consideration of other possible experimental difficulties, decide the desired accuracy of the parameters, i.e., the values of δ.
3. Calculate the value of ξ_{min} according to Eq. (14) and the values of n by Eq. (24). If $n > 5$ is obtained, carry out $n \geqq 5$ measurements. If $n > 20$, investigate the possibilities of reducing n according to Eq. (24).
4. Calculate the values of ξ_i using Eqs. (25) and (26) and those of χ_i by Eqs. (27)-(30).
5. Carry out the experiments planned, evaluate the data, and check the fulfillment of the accuracy requirement $\delta = \Delta r/r$. If the desired accuracy is not achieved, the result can be improved mainly by reducing the error of the analysis, i.e., by more careful preparation or more precise analysis of the copolymer samples. If the desired

TABLE 1. Abscissa (ξ_i) and Monomer Mixture Composition Values (χ_i mole fraction of Component 2) in the Cases of Different r_1 and r_2 Parameters ($n = 7$, $\Delta = 0.015$)

r_1	r_2	α	χ_1 ξ_1	χ_2 ξ_2	χ_3 ξ_3	χ_4 ξ_4	χ_5 ξ_5	χ_6 ξ_6	χ_7 ξ_7
0.01	0.01	1	0.9203	0.6886	0.5851	0.5000	0.4149	0.3114	0.0797
			0.0083	0.1722	0.3361	0.5000	0.6639	0.8278	0.9917
0.01	0.1	10	0.7850	0.4150	0.3097	0.2403	0.1822	0.1235	0.0267
			0.0101	0.1734	0.3367	0.5000	0.6633	0.8266	0.9899
0.01	1	100	0.5359	0.1899	0.1263	0.0909	0.0647	0.0409	0.0086
			0.0158	0.1772	0.3386	0.5000	0.6614	0.8228	0.9842
0.01	10	1000	0.2675	0.0776	0.0462	0.0307	0.0202	0.0117	0.0027
			0.0328	0.1886	0.3443	0.5000	0.6557	0.8114	0.9672
0.1	0.1	1	0.9203	0.7010	0.5912	0.5000	0.4088	0.2990	0.0797
			0.0158	0.1772	0.3386	0.5000	0.6614	0.8228	0.9842
0.1	1	10	0.7850	0.4570	0.3261	0.2403	0.1712	0.1062	0.0267
			0.0328	0.1886	0.3443	0.5000	0.6557	0.8114	0.9672
0.1	10	100	0.5359	0.2620	0.1510	0.0909	0.0532	0.0274	0.0086
			0.0797	0.2198	0.3599	0.5000	0.6401	0.7802	0.9203
1	1	1	0.9203	0.7802	0.6401	0.5000	0.3599	0.2198	0.0797
			0.0797	0.2198	0.3599	0.5000	0.6401	0.7802	0.9203
1	10	10	0.7850	0.6202	0.4275	0.2403	0.1181	0.0577	0.0267
			0.1809	0.2873	0.3936	0.5000	0.6064	0.7127	0.8191
10	10	1	0.9203	0.8687	0.7591	0.5000	0.2409	0.1313	0.0797
			0.3189	0.3792	0.4396	0.5000	0.5604	0.6208	0.6811

accuracy is not achieved even by these techniques, analogously planned experiments should be carried out to increase the number of samples.

"EFFICIENCY FACTOR" OF PLANNING

During the almost four decades that have passed since the derivation of the copolymerization composition equation, investigations of many thousands of binary copolymerization systems have been published. Nearly all these examinations were made without any planning. Nevertheless, our detailed retrospective studies [4, 5, 10] showed that a considerable part of the material published in the literature contains valuable information. Critical reexamination of existing copolymerization literature and the obtaining of reliable parameters upon reevaluation of the literature is of basic scientific interest.

For subsequent judgment on how the published data can be used for the purposes of the above reevaluation in spite of their lack of previous planning, an objective criterion is needed in addition to knowledge of r_1 and r_2, Δr_1 and Δr_2. For this purpose, we have deemed it advisable to introduce the "efficiency factor" of experimental design in order to estimate how an experimentalist had deviated from the optimal composition region. This circumstance is revealed by a comparison of the value of α according to Eq. (13) with the theoretically calculated $\alpha = r_2/r_1$ value:

$$Q = \exp\left\{ - \left| \ln \frac{r_2/r_1}{\alpha} \right| \right\} \tag{31}$$

i.e., Q equals the quotient $(r_2/r_1)/\alpha$ if it is smaller than 1, and its reciprocal value if it is higher than 1.

The better the marking out of the composition region has succeeded, the nearer is the Q value to 1. If, however, α deviates from r_2/r_1 in any direction, Q decreases and, in the limiting case, approaches zero.

Reexamination of the data of the binary copolymerization system β-propiolactone/3,3-chloromethyl-oxetane/$BF_3 \cdot Et_2O$/$MeCl_2$/-50° [11] will serve as an example of a retrospective evaluation. Figure 2 shows the KT transformation of the original experimental data, with the α value calculated by Eq. (13) ($\alpha = 64.4$). Figure 2 also contains the contours corresponding to the ±1.5% analytical error ($\Delta = 0.015$). It can be seen that the error contours are highly asymmetrical, clearly indicating the improper planning of the experiment. As a consequence, the relative error of r_1 is nearly one order of magnitude higher than that of r_2.

According to the above treatment, the correct value of α can be calculated by Eq. (12) ($\alpha = 604$). The transformation shown in Fig. 3 has been made by using this value. The error contours have become

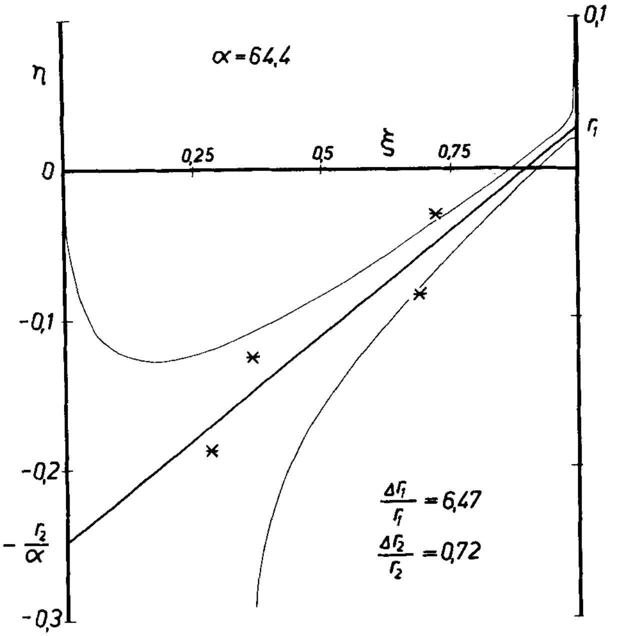

FIG. 2. KT diagram of β-propiolactone/3,3-chloromethyl-oxetane/$BF_3 \cdot Et_2O/MeCl_2$/-50°C system [11]. The error contour corresponds to the analytical error $\Delta = \pm 0.03$.

symmetrical. Improper planning is indicated by the extreme location of the original experimental points. The marks in Fig. 3 indicate the ξ_i values (n = 8) obtained from the planning proposed by us if the desired accuracy is $\delta^2 = 0.1$. The deficiencies of the original planning (Q = 0.107) are also demonstrated by the fact that two data points are below ξ_{min}.

The locations of the original and the properly planned compositions in the usual mole fraction figure of binary copolymerization are shown in Fig. 4. The curve indicates the copolymer composition calculated corresponding to the determined r_1 and r_2 values; the original data are denoted with asterisks. The composition coordinates of the planned samples are denoted with lines (the position of the 8th point is denoted with an arrow). It can be seen that they are symmetrically arranged around the polymer composition point $p_1/(p_1 + p_2) = 0.5$.

Such an arrangement of the points can also be theoretically obtained in the case of proper planning of experiments. According to Eq. (13), α is the geometrical mean value of the quantities $F = (m_1^2/m_2^2)(p_2/p_1)$. On the other hand, the planning defines α by Eq. (12), and if both α values are equal, then

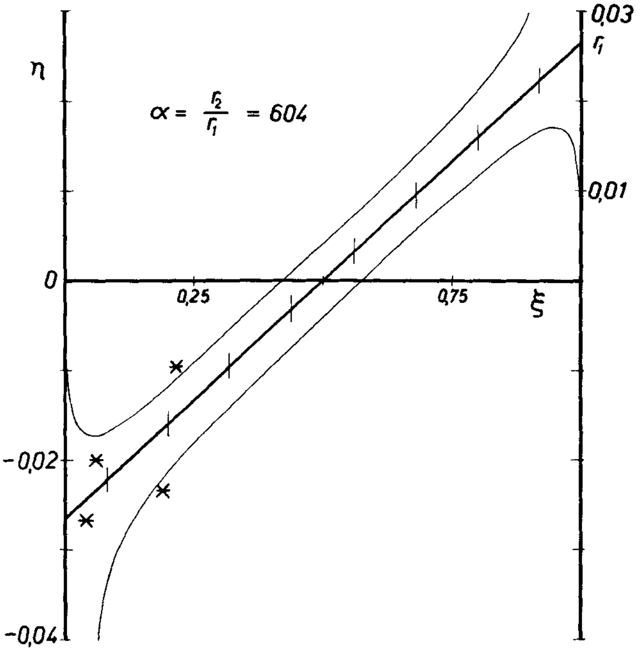

FIG. 3. Experimental data of the same system calculated with the theoretical α value ($\alpha = r_2/r_1$) ($\Delta = 0.03$). The little vertical lines indicate the ξ_i values ($n = 8$) corresponding to the $\delta^2 = 0.1$ accuracy requirement.

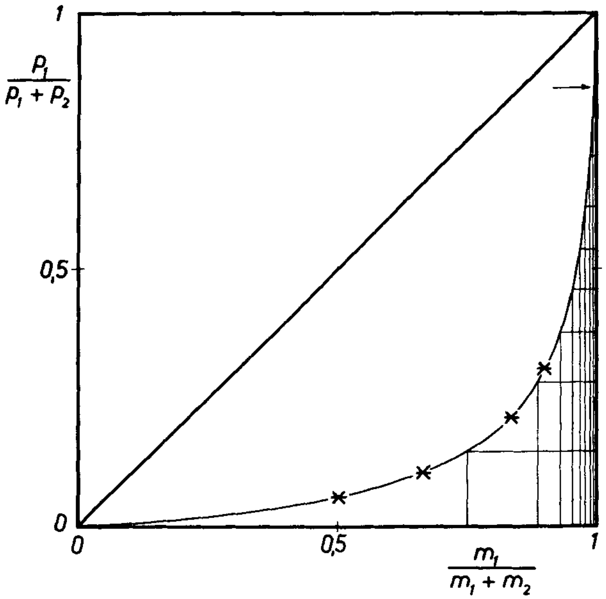

FIG. 4. Arrangements of the original and the properly planned compositions in the mole fraction figure of the binary copolymerization system studied.

$$\frac{r_2}{r_1} = \frac{\bar{m}_1^{\,2}}{\bar{m}_2^{\,2}}\,\frac{\bar{p}_2}{\bar{p}_1} \tag{32}$$

According to the copolymer composition equation,

$$\frac{\bar{p}_1}{\bar{p}_2} = \frac{\bar{m}_1}{\bar{m}_2}\,\frac{r_1\bar{m}_1 + \bar{m}_2}{\bar{m}_1 + r_2\bar{m}_2} \tag{33}$$

By comparison of these equations, in this case

$$\frac{\bar{p}_1}{\bar{p}_1 + \bar{p}_2} = 0.5 \tag{34}$$

This result indicates that a series of experiments cannot be too badly arranged, even without thorough planning, if the monomer compositions are chosen so that the copolymer composition is arranged nearly symmetrically around the $p_1/(p_1 + p_2) = 0.5$ value in the mole fraction representation. Naturally, the application of this "rule of thumb" requires certain preliminary information; namely, approximate knowledge of the mole fraction curve of copolymer composition.

REFERENCES

[1] T. Kelen and F. Tüdős, J. Macromol. Sci.–Chem., A9, 1-27 (1975); Kem. Kozl., 43, 87-111 (1975); React. Kinet. Catal. Lett., 1, 487-492 (1974).

[2] F. Tüdős, T. Kelen, T. Földes-Berezsnich, and B. Turcsânyi, Kem. Kozl., 45, 401-423 (1975); J. Macromol. Sci.–Chem., A10, 1513-1540 (1976).

[3] T. Kelen, F. Tüdős, and B. Turcsányi, Polym. Bull., 2, 71-75 (1980).

[4] J. P. Kennedy, T. Kelen, and F. Tüdős, Kem. Kozl., 44, 53-67 (1975); J. Polym. Sci., Polym. Chem. Ed., A13, 2277-2289 (1975).

[5] T. Kelen, F. Tüdős, B. Turcsányi, and J. P. Kennedy, Kem. Kozl., 46, 51-75 (1976); J. Polym. Sci., Polym. Chem. Ed., 15, 3047-3074 (1977).

[6] D. W. Behnken, J. Polym. Sci. Part A, 2, 645-668 (1964).

[7] P. W. Tidwell and G. A. Mortimer, Ibid., Part A, 3, 369-387 (1965).

[8] P. W. Tidwell and G. A. Mortimer, J. Macromol. Sci.–Rev. Macromol. Chem., C4, 281-312 (1970).

[9] M. Fineman and S. D. Ross, J. Polym. Sci., 5, 259 (1950).

[10] F. Tüdős, T. Kelen, B. Turcsânyi, and J. P. Kennedy, 5th International Symposium on Cationic and Other Ionic Polymerization, Preprints, Kyoto, 1980, pp. 171-174.

[11] K. Tada, T. Seagusa, and J. Furukawa, Makromol. Chem., 71, 71 (1964).

Accepted by editor October 8, 1980
Received for publication December 8, 1980