Multiple coordinates of the figurative points of the concentration formations of multicomponent systems*

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Received 8 January 1974

The method of multiple concentration coordinates was elaborated which renders it possible to obtain a relative change of the scale of an arbitrary part of the concentration formation of a k-component system. The centre of gravity rule (the lever rule) was proved to be valid also in the system with the transformed points.

In the presentation of a binary system ($k = 2$), the concentrations of components are presented as abscissae, either in percents or in concentration fractions. For the presentation of the concentration relations in more complicated systems, most frequently an equilateral triangle (for $k = 3$), a regular tetrahedron (for $k = 4$) or, generally for a k-component system, a regular $(k - 1)$-dimensional polyhedron are used.

If there is one or more components distinctly predominating in a part of the diagram, this part is so markedly shifted towards the figurative point of the dominating component (or components) that the diagram becomes useless for the practical purposes.

This problem was dealt with in a series of papers oriented mostly on the ternary systems. The isobaro-isothermic phase diagrams of the aqueous systems that, besides water, contain two substances with a limited and mutually comparable solubility in water under the given conditions, are most frequently represented using the rectangular system of the concentration axes, and the point of their intersection corresponds to the figurative point of pure water. Concentrations of other components are expressed as their quantities related to one and the same quantity of water. This is the so-called Schreinemakers' [1, 2] or van't Hoff's [3] method. This method, however, has some serious drawbacks. Among others, it is possible to present only a limited part of the diagram, further it cannot be used if there is a great difference between the solubilities of both substances, and the figurative points of pure substances — and consequently the corresponding binary system — are in infinity. This makes it impossible to use the centre of gravity rule (the lever rule); the validity of the rule of conodes, however, remains intact. Also the method proposed by Jänecke [4] has similar imperfections. A different treatment based upon the presentation of concentrations of non-aqueous components of a ternary system in a root scale was proposed by *Drátovský*

* Presented at the 2nd Czechoslovak Seminar on "Molten Salt Systems", Bratislava, April 11—12, 1973.

and *Ternbach* [2]. In this case, however, the straight lines are generally deformed to curves so that neither the centre of gravity rule nor the rule of conodes is valid.

The discussed problem can be solved quite generally and for all types of the systems by application of the multiple concentration units for the presentation of the composition of the system.

The multiple molecular weights were used already by Jänecke [5] for the presentation of reciprocal systems of the type

$$2MA + NB_2 \rightleftarrows 2MB + NA_2,$$

where M^{z+}, N^{2z+} are cations and A^{z-}, B^{z-} anions. This presentation was stipulated entirely by the requirement of the equality of the equivalents of the reacting substances. Analogous is the case when a nonreciprocal system, *e.g.* that consisting of the components MA, NA_2, PA_3, is presented in the coordinates 6(MA), 3(NA_2), 2(PA_3), or when a system is presented alternatively in mole and weight percents. This transformation results in a shift of all formations of the diagram.

As far as we know, the method of the multiple concentration units with the aim of enlarging some formations was for the first time purposely applied in the study of the system $Na_3AlF_6-AlF_3-Al_2O_3-CaF_2$ [6]. Here, in the plane section $2.75NaF \cdot AlF_3-Al_2O_3-CaF_2$, the concentration triangle was presented in the coordinates ($11NaF \cdot 4AlF_3$), (Al_2O_3), (CaF_2) in order to enlarge the field of the primary crystallization of cryolite, which is most important with respect to the technical practice, at the expense of the fields of the primary crystallization of Al_2O_3 and CaF_2.

The displacement of the individual parts of the concentration formation, *e.g.* of the binary system A—B, results from the fact that any arbitrary point M (a, b) of this system, if presented in the coordinates (f_A A), (B) ($f_A \neq 1$), geometrically approaches the point B for $f_A > 1$, and the point A for $f_A < 1$.

The proposed method makes it easy to find such a multiple of the concentration unit of components of a given system that any arbitrary point M, which is inside the concentration formation of the system can be transferred to another arbitrary point M′ of the same concentration formation.

1. Denotation and symbols

The initial coordinates of the point M (it holds for both the mole and the weight % as well as for the mole and weight concentration fractions): a % of component A, b % of component B, ..., l % of component L.

The required coordinates of the point M′: a' % of component A, b' % of component B, ..., l' % of component L.

The multiple of the individual concentration units will be denoted as the *multiplying factor*. The shift (transformation) of the point M (a, b, ..., l) into M′ (a', b', ..., l') will be denoted as

$$M\,(a, b, \ldots, l) \rightarrow M'\,(a', b', \ldots, l'). \qquad (1)$$

The points M and M′ for which holds (*1*) will be denoted as *associated points*.

2. Transformations in a binary system

2.1. Algebraic treatment of the problem

The transformed coordinates are introduced in the following way

$$a + b = \frac{a}{f_A} \cdot f_A + \frac{b}{f_B} \cdot f_B = m f_A + n f_B .$$

It should be kept in mind that $a + b = 1$ (or 100), however $m + n \neq 1$ (or 100). For the transformed coordinates a', b' $(a' + b' = 1)$ it holds

$$a' : b' = m : n .$$

Evidently $m = a/f_A$; $n = b/f_B$; $m/n = a f_B/b f_A$.

$$f_A = \frac{a}{b} \cdot \frac{n}{m} \cdot f_B = \frac{a}{b} \cdot \frac{b'}{a'} \cdot f_B , \tag{2a}$$

$$F = \frac{f_A}{f_B} = \frac{a}{b} \cdot \frac{b'}{a'} . \tag{2b}$$

For the normalizing coefficient $k(m + n) = 1$ it holds

$$k = a'/m = b'/n .$$

Regarding the validity of the relations $a + b = 1$, $a' + b' = 1$, from (2b) it follows:

$$a' = \frac{a}{F - a(F - 1)} , \tag{3a}$$

$$b' = \frac{b F}{1 + b(F - 1)} . \tag{3b}$$

In the solution of the eqns (2a), (2b), one of the factors f_A, f_B can be chosen arbitrarily. From the practical standpoint it appears to be advantageous to choose min $(f_A, f_B) = 1$. Then the transformed coordinates a', b' can be easily determined by means of eqn (2a) or (2b).

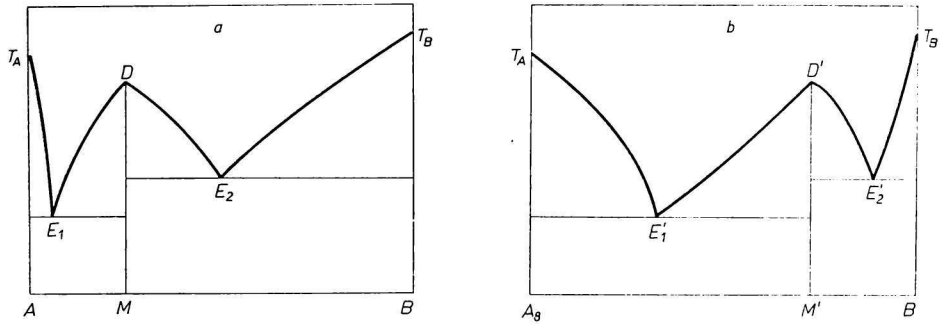

Fig. 1. The isobaric phase diagram of the binary system A—B with a congruently melting compound M ≡ A_3B: *a*) in coordinates (A, B); *b*) in coordinates (A_8, B).

Example: In the system A—B (Fig. 1*a*), there exists the compound M ≡ A_3B, which forms simple eutectic subsystems with the components A and B. Let be E_1: 94.3% A, 5.7% B; E_2: 50.0% A, 50.0% B. The multiplying factor is to be assessed in such a way that the point E_1 will be transferred into 1/3 of the distance between A and B.

Evidently $a_1 = 94.3$, $b_1 = 5.7$; $a_1' = 66.6$, $b_1' = 33.3$;

$$f_A = \frac{94.3}{5.7} \cdot \frac{33.3}{66.6} \cdot f_B = 8.2719 f_B .$$

We put $f_B = 1$ and round the factor f_A to $8 = F$. Then the system is presented in coordinates (8A, B) = (A_8, B) and the coordinates of the characteristic points are:

$$E_1: \frac{b_1'}{a_1'} = F \frac{b_1}{a_1} = 8 \frac{5.7}{94.3} = 0.48356 .$$

From there $a_1' = 67.405\%$ A_8, $b_1' = 32.595\%$ B. By analogy, we determine:

$$M: a_1' = 27.273\% A_8, \; b_1' = 72.727\% B,$$

$$E_2: a_1' = 11.\bar{1}\% A_8, \; b_1' = 88.\bar{8}\% B .$$

From the comparison of Figs. 1*a* and 1*b* it is evident that in the process of the transformation of the concentration coordinates some liquidus curves (initially concave) become convex with respect to the concentration axis. In [7] the condition for the occurrence of the convex parts on the liquidus curves was found to be the inequality $\Delta S_i^f < 2R$ (where ΔS_i^f is the entropy of fusion of the component i and R is the gas constant). Evidently, this condition holds in the presentation of the phase diagram in simple (not multiple) coordinates. This problem will be dealt with in a subsequent paper.

2.2. Transformation of coordinates in the conversion from weight to mole percents

Let us consider the figurative point M in a binary system A—B with the coordinates a weight % of component A, b weight % of component B. If the composition of the point M is to be converted into the mole %, for the new coordinates a', b' the following relations are obtained:

$$a' = \frac{100a}{M_A} : \left[\frac{a}{M_A} + \frac{b}{M_B}\right]; \qquad b' = \frac{100b}{M_B} : \left[\frac{a}{M_A} + \frac{b}{M_B}\right],$$

where M_A and M_B are the molecular weights of the substances A and B, respectively.

After substituting for a' and b' in the eqn (*2b*) and rearranging, for the conversion from weight to mole percents the following relation is obtained:

$$F_{wt \to mole} = f_A : f_B = M_A : M_B . \qquad (4)$$

If $M_A > M_B$, then $a' < a$, which means that the point M is shifted from A towards B, and reversely for $M_A < M_B$.

Therefore, if the multiplying factor equals the ratio $M_A : M_B$, the formula for the conversion from weight % to mole % is obtained as a particular case of the method of the multiple coordinates.

Reversely, if a = mole % A, b = mole % B, then

$$F_{mole \to wt} = M_B/M_A , \qquad (5)$$

or

$$F_{mole \to wt} = F^{-1}_{wt \to mole} . \qquad (6)$$

2.3. *Graphic solution of the problem*

Though the calculation of the multiplying factor and of the transformed coordinates of components is simple, the graphic method of determination of new coordinates appears to be more convenient when a larger group of points is involved. This method is convenient especially in such a case when for the ratio of the both multiplying factors the inequality is valid:

$$\frac{\max(f_A, f_B)}{\min(f_A, f_B)} < 10. \qquad (7)$$

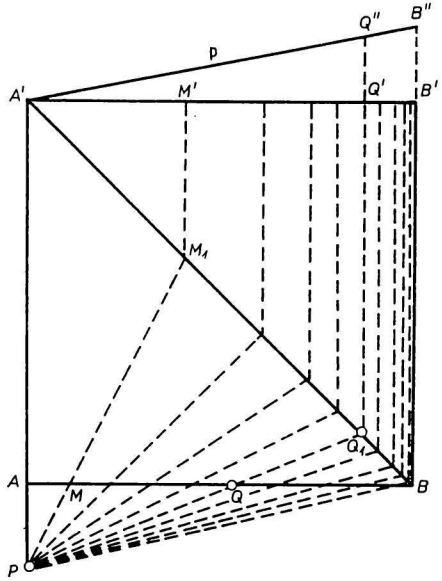

Fig. 2. The graphic method for determining of the coordinates of the transformed points of the system A′—B′ and of the multiplying factor (for $F = 6$).

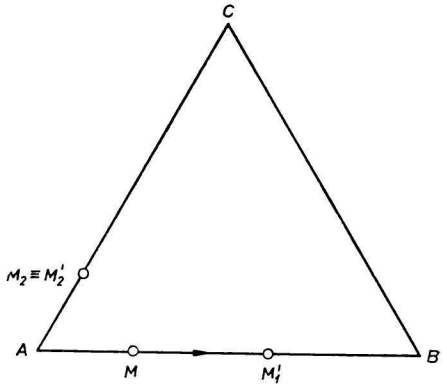

Fig. 3. The shift of the point M_1 situated at the side AB of the concentration triangle of the ternary system A—B—C into the position M_1'. The point M_2, situated at the side AC of the same concentration triangle, keeps its place.

The graphic proceeding is presented in Fig. 2. For the known factor f_A, the square ABB′A′ is drawn where A—B is the initial system and A′—B′ is the system with the transferred points. (Instead of a square also rectangle or a parallelogram can be used on condition that the couple of associated points M and M′ will be placed on the two parallel sides corresponding to the systems A—B and A′—B′.)

The abscissa A′B′ is divided in proportion $f_A : 1$, *e.g.* in such a way that on the half-straight line p with origin in A′ in an arbitrary scale the distances $A'Q'' = f_A$ and $Q''B'' = 1$ are marked and the point Q″ is projected to A′B′ by a ray which is parallel with B″B′. Thus the point Q′ is obtained through which a parallel with AA′ is drawn to the intersection with the diagonal A′B. The junction of this intersection (point Q_1) with the point Q which halves the abscissa AB, intersects the prolonged abscissa A′A

in the point P. This point is a pole from which the points M_i of the initial system A—B are projected into the transferred positions M_i' in a way which is obvious from the diagram.

If a couple of associated points (M, M′) is given, one proceeds in the following way: a parallel with A′A is drawn through the point M′ to the intersection with the diagonal A′B (point M_1). The junction M_1M crosses the straight line A′A in the point P. The subsequent proceeding is analogous to the previous case. The value of f_A is determined graphically in such a way that the point Q, which halves the distance between A and B, is projected from the pole P to the diagonal A′B and from there to the abscissa A′B′ (point Q′). From eqn (*2a*) for $a = b = 50$, $f_B = 1$ it follows that $A'Q' : Q'B' = f_A : 1$.

If $f_B \neq 1$, the above proceeding remains preserved, the only difference being that instead of the factor f_A the ratio $f_A/f_B = F$ is used in all relations.

The proceeding presented in Fig. 2 can be used also in conversion from the weight to mole percents and *vice versa*. In this case, F is determined by means of the relations (*4*) or (*5*).

The correctness of the graphic proceeding for the determination of transformed coordinates can be easily proved also analytically.

2.4. Centre of gravity rule in a transformed system

For the initial system in coordinates (A, B) it holds

$$[n_A^{(1)} + n_B^{(1)}]\, a_1 + [n_A^{(3)} + n_B^{(3)}]\, a_3 = [n_A^{(1)} + n_B^{(1)} + n_A^{(3)} + n_B^{(3)}]\, a_2,$$

where $n_{A,B}^{(i)}$ is the number of moles of the substances A and B in the systems "1" and "3"; a_1 and a_3 is the content of the substance A in the respective systems expressed in concentration fractions or in percents, a_2 is the content of the substance A in the resulting system "2".

After rearranging, the well-known relation is obtained:

$$\frac{n_A^{(1)} + n_B^{(1)}}{n_A^{(3)} + n_B^{(3)}} = \frac{a_2 - a_3}{a_1 - a_2}. \tag{8}$$

For the transformed system in coordinates (f_A A, f_B B), whilst $f_A/f_B = F$, it holds analogously:

$$[n_A^{(1)}/F + n_B^{(1)}]\, a_1' + [n_A^{(3)}/F + n_B^{(3)}]\, a_3' = [n_A^{(1)}/F + n_B^{(1)} + n_A^{(3)}/F + n_B^{(3)}]\, a_2'.$$

From there:

$$[n_A^{(1)}/F + n_B^{(1)}] : [n_A^{(3)}/F + n_B^{(3)}] = (a_2' - a_3') : (a_1' - a_2'). \tag{9}$$

It is to be proved that the relation (*8*) identically equals the relation (*9*).

For the left side of eqn (*9*) it is evidently valid:

$$[n_A^{(1)}/F + n_B^{(1)}] : [n_A^{(3)}/F + n_B^{(3)}] = [n_A^{(1)} + F\, n_B^{(1)}] : [n_A^{(3)} + F\, n_B^{(3)}] =$$

$$= \left\{[n_A^{(1)} + n_B^{(1)}]\left[\frac{n_A^{(1)}}{n_A^{(1)} + n_B^{(1)}} + F\,\frac{n_B^{(1)}}{n_A^{(1)} + n_B^{(1)}}\right]\right\} \cdot$$

$$\cdot \left\{[n_A^{(3)} + n_B^{(3)}]\left[\frac{n_A^{(3)}}{n_A^{(3)} + n_B^{(3)}} + F\,\frac{n_B^{(3)}}{n_A^{(3)} + n_B^{(3)}}\right]\right\}^{-1} \cdot$$

Since

$$\frac{n_A^{(i)}}{n_A^{(i)} + n_B^{(i)}} = a_i, \quad \frac{n_B^{(i)}}{n_A^{(i)} + n_B^{(i)}} = b_i, \quad a_i + b_i = 1$$

then

$$[n_A^{(1)}/F + n_B^{(1)}] : [n_A^{(3)}/F + n_B^{(3)}] = \frac{n_A^{(1)} + n_B^{(1)}}{n_A^{(3)} + n_B^{(3)}} \cdot \frac{F - a_1(F-1)}{F - a_3(F-1)} . \qquad (10)$$

The right side of eqn (9) is rearranged using the relation (3a), *i.e.* $a_i' = a_i[F - a_i(F-1)]^{-1}$. Then it holds

$$\frac{a_2' - a_3'}{a_1' - a_2'} = \frac{a_2 - a_3}{a_1 - a_2} \cdot \frac{F - a_1(F-1)}{F - a_2(F-1)} . \qquad (11)$$

After introducing from eqns (10) and (11) into eqn (9) the relation (8) is obtained. Thus the identity of eqns (8) and (9) has been proved.

It is thus evident that with this method of transformation of the coordinates the validity of the centre of gravity rule (the lever rule) is preserved, this being one of the main advantages of the proposed method.

3. Transformations in a ternary system

3.1. *Points which are to be transformed are on one side of the concentration triangle, e.g. AB* (Fig. 3)

The required transformation:

$$M_1 (a_1, b_1) \rightarrow M_1' (a_1', b_1').$$

The points on the side AC remain in their places:

$$M_2 (a_2, c_2) \rightarrow M_2' (a_2, c_2); \quad M_2 \equiv M_2'.$$

For $M_1 \rightarrow M_1'$ it holds

$$f_A^{(1)} = a_1 b_1' f_B / a_1' b_1 .$$

For $M_2 \rightarrow M_2'$ it holds

$$f_A^{(2)} = a_2 c_2 f_C / a_2 c_2 = f_C .$$

With regard to the fact that in a definite system for the component A the same multiple, which is valid both in the subsystem A–B and in the subsystem A–C, has to be used, it holds that $f_A^{(1)} = f_A^{(2)}$. If we put $f_C = 1$, then also $f_A = 1$ and the concentration triangle is to be presented in the coordinates $(f_A\,A, f_B\,B, f_C\,C) = \left(A, \frac{a_1' b_1}{a_1 b_1'} B, C\right)$.

Example:

$$a_1 = 0.8, \; b_1 = 0.2; \quad a_1' = 0.5, \; b_1' = 0.5.$$

$$f_A = a_1 b_1' f_B / a_1' b_1 = 4 f_B = f_C; \quad \text{let be } f_C = f_A = 1.$$

Then $f_B = 1/4$ and the concentration triangle is to be presented in the coordinates (A, B/4, C); or $f_B = 1$ and then the coordinates are (4A, B, 4C).

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3.2. The points, which are to be transformed are on two sides of the concentration triangle (Fig. 4)

The required transformation

$$M_1\,(a_1, b_1) \rightarrow M_1'\,(a_1', b_1'); \quad M_2\,(a_2, c_2) \rightarrow M_2'\,(a_2', c_2').$$

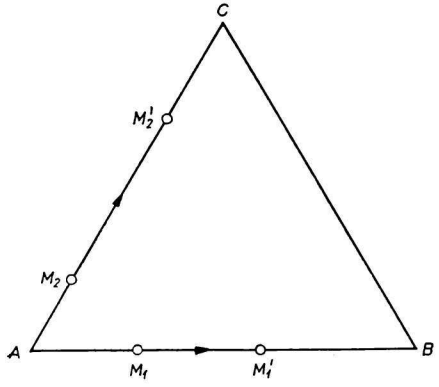

Fig. 4. The shift of points situated on the two sides of the concentration triangle of the ternary system A—B—C. The point M_1 is to be shifted into the position M_1', the point M_2 into the position M_2'.

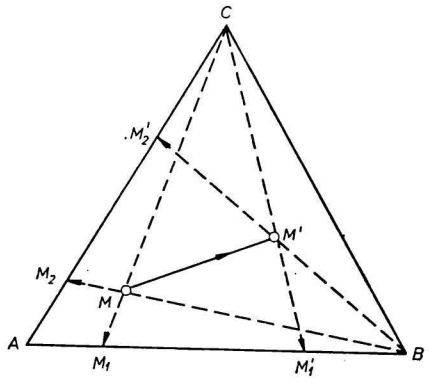

Fig. 5. The shift of the point M, situated inside the concentration triangle of the ternary system A—B—C, into the position M'.

In the first step, the factor F is to be determined:

$$f_A^{(1)} = \frac{a_1}{b_1} \cdot \frac{b_1'}{a_1'} \cdot f_B; \quad f_A^{(2)} = \frac{a_2}{c_2} \cdot \frac{c_2'}{a_2'} \cdot f_C;$$

evidently $f_A^{(1)} = f_A^{(2)}$. Then it holds

$$f_B = \frac{a_2}{c_2} \cdot \frac{c_2'}{a_2'} \cdot \frac{b_1}{a_1} \cdot \frac{a_1'}{b_1'} \cdot f_C.$$

It is advantageous to put min $(f_A, f_B, f_C) = 1$. Let it be f_C. Then it holds

$$f_A = \frac{a_2}{a_2'} \cdot \frac{c_2'}{c_2}; \quad f_B = \frac{a_1'}{a_1} \cdot \frac{a_2}{a_2'} \cdot \frac{b_1}{b_1'} \cdot \frac{c_2'}{c_2}.$$

The required concentration coordinates are:

$$\left[\frac{a_2}{a_2'} \cdot \frac{c_2'}{c_2} \cdot A, \quad \frac{a_1'}{a_1} \cdot \frac{a_2}{a_2'} \cdot \frac{b_1}{b_1'} \cdot \frac{c_2'}{c_2} \cdot B, C\right],$$

Example:

$$a_1 = 0.8,\ b_1 = 0.2; \quad a_1' = 0.5,\ b_1' = 0.5.$$

$$a_2 = 0.6,\ c_2 = 0.4; \quad a_2' = 0.2,\ c_2' = 0.8.$$

$$f_A^{(1)} = 0.8 \cdot 0.5 \cdot f_B / 0.2 \cdot 0.5 = 4 f_B,$$

$$f_A^{(2)} = 0.6 \cdot 0.8 \cdot f_C / 0.4 \cdot 0.2 = 6 f_C .$$

If we put $f_C = 1$, then the required coordinates are (6A, 3B/2, C). If we put $f_C = 2$, the coordinates are (12A, 3B, 2C).

It is evident that the transformation of the points situated on the sides of the concentration triangle is accompanied also by a shift of points inside the concentration triangle.

3.3. The points, which are to be transformed are situated inside the concentration triangle

The proceeding in the transformation of the concentration coordinates of the point M (a, b, c) to the coordinates M′ (a', b', c') (Fig. 5) is as follows:

The point M is projected from the vertex C to the side AB (in this way the point M_1 is obtained), as well as from the vertex B on the side AC (thus the point M_2 is obtained). The points M_1, M_2 are shifted into the positions M_1', M_2' in such a way that M_1' should be identical with the projection of the point M′ to the side AB and M_2' with the projection of the point M′ to the side AC.

The coordinates of the auxiliary points are:

$$M_1 (a_1, b_1) \rightarrow M_1' (a_1', b_1'),$$

$$M_2 (a_2, c_2) \rightarrow M_2' (a_2', c_2').$$

For the multiplying factors it holds

$$f_A^{(1)} = \frac{a_1}{b_1} \cdot \frac{b_1'}{a_1'} \cdot f_B ,$$

$$f_A^{(2)} = \frac{a_2}{c_2} \cdot \frac{c_2'}{a_2'} \cdot f_C .$$

From the equality $f_A^{(1)} = f_B^{(2)}$ it follows:

$$f_B = \frac{a_2}{c_2} \cdot \frac{c_2'}{a_2'} \cdot \frac{b_1}{a_1} \cdot \frac{a_1'}{b_1'} \cdot f_C .$$

For the coordinates of the points M, M′, M_1, M_1', M_2, M_2' the following relations are valid:

$$a_1 : b_1 = a : b, \quad a_1' : b_1' = a' : b';$$

$$a_2 : c_2 = a : c, \quad a_2' : c_2' = a' : c'.$$

Consequently, the expression for f_B is simplified:

$$f_B = \frac{b}{b'} \cdot \frac{c'}{c} \cdot f_C .$$

Let be $\min (f_A, f_B, f_C) = f_C = 1$. Then the components of the concentration triangle can be expressed in units:

$$\left(\frac{a}{a'} \cdot \frac{c'}{c} \cdot \mathrm{A}, \frac{b}{b'} \cdot \frac{c'}{c} \cdot \mathrm{B}, \mathrm{C} \right)$$

or

$$\left(\frac{a}{a'} \cdot \mathrm{A}, \frac{b}{b'} \cdot \mathrm{B}, \frac{c}{c'} \cdot \mathrm{C}\right).$$

The transformation of the coordinates of the formations inside the concentration triangle is evidently accompanied by a transformation of points which are situated on the sides of the triangle.

4. Transformations in a *k*-component system

The case will be dealt with when the point, which is to be shifted is inside the concentration polyhedron of the system. The transformation is to be:

$$\mathrm{M}\,(a, b, c, \ldots, k) \rightarrow \mathrm{M}'\,(a', b', c', \ldots, k').$$

By generalizing the consideration from the paragraph 3.3. and putting $f_{\mathrm{K}} = 1$, for a k-component system the following relations are obtained:

$$f_{\mathrm{A}} = \frac{a\,k'}{a'\,k}, \quad f_{\mathrm{B}} = \frac{b\,k'}{b'\,k}, \quad f_{\mathrm{C}} = \frac{c\,k'}{c'\,k}, \quad \ldots\,.$$

Then the components of the concentration polyhedron are expressed as:

$$(f_{\mathrm{A}}\,\mathrm{A}, f_{\mathrm{B}}\,\mathrm{B}, f_{\mathrm{C}}\,\mathrm{C}, \ldots, \mathrm{K}) = \frac{a\,k'}{a'\,k} \cdot \mathrm{A}, \frac{b\,k'}{b'\,k} \cdot \mathrm{B}, \ldots, \mathrm{K}.$$

If we put $f_{\mathrm{K}} = k/k'$, for the coordinates of polyhedron the relation is obtained:

$$\left(\frac{a}{a'} \cdot \mathrm{A}, \frac{b}{b'} \cdot \mathrm{B}, \ldots, \frac{k}{k'} \cdot \mathrm{K}\right).$$

An analogous proceeding can be used also in the case when the concentrations are given in weight percents. Then the weight concentration units for particular components generally differ from each other.

In the above proceeding, the centre of gravity rule (the lever rule) is preserved.

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Translated by K. Matiašovský