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CRYSTALLOGRAPHIC SOFTWARE

# Triangulation of Salt Systems with Barium Borate

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**Abstract**—A program has been developed that implies the following operations: vertices of an $n$-component system are presented in the form of a graph, the adjacency matrix and the adjacency list with zero adjacency-matrix elements are written, the elements of the adjacency list are multiplied taking into account the absorption rule, and inversion is performed. The local $X(x_1, ..., x_m)$ barycentric coordinate subsystems in the unified space of an $n$-component system $Z(z_1, ..., z_n)$ are related by the equation $Z = KX$, where the $Z$ coordinates of subsystem vertices are located in the columns of the matrix $K$ $[m \times n]$. The membership of the composition $G(g_1, ..., g_n)$ in subsystem $K$ is determined by the condition $0 < x_i < 1$ for the roots of the equation $G = KX$. When recognizing the simplices formed by internal vertices, polyhedration is performed twice: first without these points and then only with respect to the microcomplexes with internal points. Formulas that relate the number of obtained simplices to the adjacency matrix have been derived to verify the results of polyhedration.

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## INTRODUCTION

Single crystals of the low-temperature modification of barium metaborate are widely used in nonlinear optics, in particular for laser frequency conversion in the visible and UV regions [1, 2]. To choose appropriate solvents for their growth, reciprocal salt systems (presented in the form of polyhedra) are investigated. The formation of compounds calls for the polyhedration of the concentration space (partition of complexes into simplices and (or) simpler complexes). After determining the stable cutting elements, it is necessary to identify each obtained subsystem. When speaking about identification, one should mean either the establishment of affiliation of composition to a specific subsystem or, vice versa, the possibility of finding a composition coinciding with the center of mass of material points in the vertices of this subsystem. The polyhedration of reciprocal salt systems encounters some problems. One of them is related to the existence of isolated vertices in simplices, which correspond, e.g., to the $Na_2Sr(VO_3)_4$ compound in the $Na_2O–V_2O_5–SrO$ system [3].

## FORMULAS THAT RELATE BARYCENTRIC COORDINATES

The establishment of relationship between concentration coordinates is related to the problem of identifying a point as a center of mass in different groups of material points. This class of problems also includes the determination of affiliation of a point to a simplex or a complex, as well as the problem of polyhedration of concentration complexes into simplices and microcomplexes of the same dimension [4–10].

The barycentric coordinates (B coordinates) of any subsystem $X$ of dimension $m$ can be related to the coordinates of a primitive system $Z$, which is composed of $n$ simple elements ($m \le n$), using the matrix product, in terms of the matrix $K$ of dimension ($n \times m$), the columns of which contain the $Z$ coordinates of vertices of simplex $X$:

$$Z = K \cdot X \quad \text{or} \quad \begin{pmatrix} z_1 \\ z_2 \\ \dots \\ z_n \end{pmatrix} = \begin{pmatrix} k_{11} & k_{12} & \dots & k_{1m} \\ k_{21} & k_{22} & \dots & k_{2m} \\ \dots & \dots & \dots & \dots \\ k_{n1} & k_{n2} & \dots & k_{nm} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ \dots \\ x_m \end{pmatrix}. \quad (1)$$

The concentration of an $n$-component mixture can be expressed in mass, molar, or equivalent fractions. If two systems of B coordinates, $Z$ and $Y$, have the same dimension ($n$) and coinciding coordinates of simplex vertices but differ in masses placed in these vertices and if the simplex vertices contain masses $p_i$ and $q_i$ in the systems $Z$ and $X$, respectively, the coordinates $z_i$ and $y_i$ are related as follows:

$$\frac{z_i/q_i}{\sum_{j=1}^{n}(z_j/q_j)} = \frac{y_i/p_i}{\sum_{j=1}^{n}(y_j/p_j)}, \quad i = \overline{1,n}. \quad (2)$$

Formulas (2) relate coordinates expressed in terms of equivalent fractions. If all weighting coefficients $p_i = 1$, the composition is expressed in terms of molar fractions.

The mass ($B_i$) and molar ($N_i$) fractions are related by the expressions

$$B_i = \frac{N_i M_i}{\sum_{j=1}^{n}(N_j M_j)}, \quad N_i = \frac{B_i}{M_i \sum_{j=1}^{n}(B_j/M_j)}, \qquad (3)$$

where $M_i$ is the mass of one mol of the $i$th component.

## IDENTIFICATION OF TERNARY HETERO-COMPOUND $BA_2NA_3(BO_2)_6F$ WITH RESPECT TO THE BA–NA–B–O–F SYSTEM AND ITS SUBSYSTEMS

Hetero-compound $D = Ba_2Na_3(B_3O_6)_2F$ [11,12] can be represented in the square of the ternary reciprocal system Ba,Na||$BO_2$,F; in the prism of the quaternary reciprocal fluoride-oxide system Ba,Na,B||F,O; and in the pentatop of the quinary system Ba–Na–B–O–F (Fig. 1).

In the pentatop of the quinary system Ba–Na–B–O–F, the B coordinates of the compound $D$ are [Ba] = 2, [Na] = 3, [B] = 6, [O] = 12, [F] = 1 or, after normalization, (2/24, 3/24, 6/24, 12/24, 1/24).

A transition from the B coordinates (2/24, 3/24, 6/24, 12/24, 1/24) of the compound in the quinary system Ba–Na–B–O–F to the subsystem (simplex) $1/7BaB_2O_4$–$1/3BaF_2$–$1/4$ $NaBO_2$ of the reciprocal system $1/7$ $BaB_2O_4$–$1/3$ $BaF_2$–$1/2$ NaF–$1/4$ $NaBO_2$ leads to the equation

$$\begin{pmatrix} 2/24 \\ 3/24 \\ 6/24 \\ 12/24 \\ 1/24 \end{pmatrix} = \begin{pmatrix} 1/7 & 1/3 & 0 \\ 0 & 0 & 1/4 \\ 2/7 & 0 & 1/4 \\ 4/7 & 0 & 2/4 \\ 0 & 2/3 & 0 \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix},$$

$$x_1 = \frac{7}{16}, \quad x_2 = \frac{1}{16}, \quad x_3 = \frac{8}{16},$$

(it follows from the equalities $3/24 = x_3/4$ and $1/24 = 2x_2/3$ that $x_3 = 12/24 = 8/16$ and $x_2 = 3/48 = 1/16$; hence, the normalization condition yields $x_1 = 1 - x_2 - x_3 = 7/16$).

This solution means that, to form a unit mass $D = Ba_2Na_3B_6O_{12}F$ ($q_D = 1$), one must take $p_{BaB_2O_4} = 7/16$, $p_{BaF_2} = 1/16$, amd $p_{NaBO_2} = 8/16$:

$$1/24Ba_2Na_3B_6O_{12}F = 7/16Ba_{1/7}B_{2/7}O_{4/7} + 1/16Ba_{1/3}F_{2/3} + 8/16Na_{1/4}B_{1/4}O_{2/4}.$$

Indeed, since the initial material points contained the masses

$$p_{BaB_2O_4} = p_{Ba} + p_B + p_O = 1/7 + 2/7 + 4/7 = 1,$$

$$p_{BaF_2} = p_{Ba} + m_F = 1/3 + 2/3 = 1,$$

$$p_{NaBO_2} = p_{Na} + p_B + p_O = 1/4 + 1/4 + 2/4 = 1,$$

having taken three salts of unit mass in the ratio of 7 : 1 : 8, one obtains a unit mass of the compound $D$:

$$\frac{7}{16}(p_{Ba} + p_B + p_O) + \frac{1}{16}(p_{Ba} + p_F) + \frac{8}{16}(p_{Na} + p_B + p_O)$$

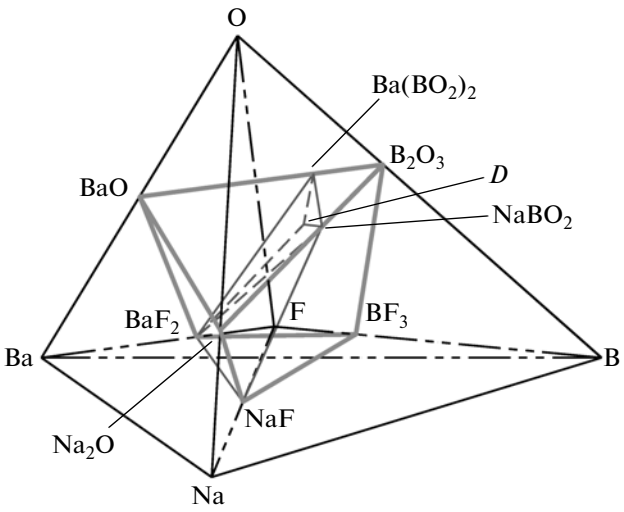


**Fig. 1.** Compound $D = Ba_2Na_3(B_3O_6)_2F$ in pentatope of quinary system Ba–Na–B–O–F.

$$= \frac{7}{16}\left(\frac{1}{7}Ba + \frac{2}{7}B + \frac{4}{7}O\right) + \frac{1}{16}\left(\frac{1}{3}Ba + \frac{2}{3}F\right) + \frac{8}{16}\left(\frac{1}{4}Na + \frac{1}{4}B + \frac{2}{4}O\right)$$

$$= \frac{2}{24}Ba + \frac{3}{24}Na + \frac{6}{24}B + \frac{12}{24}O + \frac{1}{24}F\Big)$$

$$= \frac{1}{24}Ba_2Na_3B_6O_{12}F.$$

Thus, the location of the compound $D$ in the subsystem $1/7BaB_2O_4$–$1/3BaF_2$–$1/4NaBO_2$ is described by the ratio 7 : 1 : 8.

Let us now increase the masses of salts in the square vertices so as to make the coefficients at them equal to unity (i.e., we pass to a new coordinate system by locating the masses $q_{BaB_2O_4} = 7$, $q_{BaF_2} = 3$, $q_{NaF} = 2$, and $q_{NaBO_2} = 4$ at the material points $1/7BaB_2O_4$, $1/3BaF_2$, $1/2$NaF, and $1/4NaBO_2$, respectively). Then, according to formula (2), the point of the compound $D$ with the coordinates (7/16, 1/16, 8/16) is transferred to the position with the coordinates

$$y_1 = \frac{(7/16)/7}{(7/16)/7 + (1/16)/3 + (8/16)/4} = \frac{3}{10},$$

$$y_2 = \frac{(1/16)/3}{(7/16)/7 + (1/16)/3 + (8/16)/4} = \frac{1}{10}, \quad y_3 = \frac{6}{10}$$

or to the position 3 : 1 : 6 with the mass of compound $D$ equal to 2:

$$2Ba_2Na_3B_6O_{12}F = 3BaB_2O_4 + BaF_2 + 6NaBO_2.$$

Hence, in the subsystem $BaB_2O_4$–$BaF_2$–$NaBO_2$, the compound $D$ is located at a point with the coordinates (3/10, 1/10, 6/10) and correspond to the ratio of the initial salts as 3 : 1 : 6.

If we double the masses at the vertices of sodium fluoride and sodium borate, i.e., reduce the number of anions forming the initial salt to two by forming the subsystem $Ba(BO_2)_2$–$BaF_2$–$Na_2(BO_2)_2$ in the ternary reciprocal system $Ba(BO_2)_2$–$BaF_2$–$Na_2F_2$–$Na_2(BO_2)_2$, the coordinates of point $D$ will change. This change can be determined in two ways, i.e., by passing

(i) from (7/16, 1/16, 8/16) of the subsystem $1/7Ba(BO_2)_2$–$1/3BaF_2$–$1/4NaBO_2$

$$\text{with} \quad p_{BaB_2O_4} = p_{BaF_2} = p_{NaBO_2} = 1; \quad \text{or}$$

(ii) from (3/10, 1/10, 6/10) of the subsystem $Ba(BO_2)_2$–$BaF_2$–$NaBO_2$

$$\text{with} \quad q_{BaB_2O_4} = 7, \quad q_{BaF_2} = 3, \quad q_{NaBO_2} = 4.$$

In the first case, one uses the expressions

$$y_1 = \frac{(7/16)/7}{(7/16)/7 + (1/16)/3 + (8/16)/8} = \frac{3}{7},$$

$$y_2 = \frac{1}{7}, \quad y_3 = \frac{3}{7}.$$

In the second case, it is necessary to construct the equations

$$\frac{(3/10)/7}{(3/10)/7 + (1/10)/3 + (6/10)/8} = \frac{y_1/7}{y_1/7 + y_2/3 + y_3/4},$$

$$\frac{(1/10)/3}{(3/10)/7 + (1/10)/3 + (6/10)/8} = \frac{y_2/3}{y_1/7 + y_2/3 + y_3/4},$$

the solution of which yields the same results, i.e., $y_1 = 3/7$, $y_2 = 1/7$, $y_3 = 3/7$. Therefore, when doubling the mass of the $NaBO_2$ point, in the $BaB_2O_4$–$BaF_2$–$Na_2(BO_2)_2$ subsystem, the material point $D$ with a mass of 2 is transferred to the point (3/7, 1/7, 3/7), according to the ratios of the initial salts, 3 : 1 : 3:

$$2Ba_2Na_3B_6O_{12}F = 3BaB_2O_4 + BaF_2 + 3Na_2(BO_2)_2.$$

To express a compound of a ternary reciprocal system in terms of the contents of system-forming salts, one must determine the content of each ion in the compound and then multiply pairwise the number of cations by the number of corresponding anions. The thus obtained values also become coefficients in the expansion of the compound in four salt components.

For example, in the compound $Ba_2Na_3(BO_2)_6F$ of the ternary reciprocal system $Ba(BO_2)_2$–$BaF_2$–$NaF$–$NaBO_2$ with masses in the complex vertices $q_{Ba(BO_2)_2} = 7$, $q_{BaF_2} = 3$, $q_{NaF} = 2$, and $q_{NaBO_2} = 4$, the numbers of cations are [Ba] = 2 and [Na] = 3, while the numbers of anions are $[BO_2] = 6$ and [F] = 1. Therefore, the numbers of salts will be as follows: $[Ba(BO_2)_2] = 2 \cdot 6 = 12$, $[BaF_2] = 2$, [NaF] = 3, and $[NaBO_2] = 3 \cdot 6 = 18$. As a result, the compound can be decomposed into four initial salts of the system $Ba(BO_2)_2$–$BaF_2$–$NaF$–$NaBO_2$ as 12 : 2 : 3 : 18 or

$$7Ba_2Na_3(BO_2)_6F = 12Ba(BO_2)_2 + 2BaF_2 + 3NaF + 18NaBO_2.$$

In the $Ba_2Na_3(BO_2)_6F$ compound, in the $Ba(BO_2)_2$–$BaF_2$–$Na_2F_2$–$Na_2(BO_2)_2$ system with masses of $q_{Ba(BO_2)_2} = 7$, $q_{BaF_2} = 3$, $q_{Na_2F_2} = 4$, and $q_{Na_2(BO_2)} = 8$, there are [Ba] = 2 and $[Na_2] = 3/2$ cations and $[(BO_2)_2] = 3$ and $[F_2] = 1/2$ anions. Thus, the numbers of the salts will be $[Ba(BO_2)_2] = 2 \cdot 3 = 6$, $[BaF_2] = 2 \cdot (1/2) = 1$, $[Na_2F_2] = (3/2) \cdot (1/2) = 3/4$, and $[Na_2(BO_2)_2] = (3/2) \cdot 3 = 9/2$, and the compound under consideration can be expanded into four initial salts of the system $Ba(BO_2)_2$–$BaF_2$–$Na_2F_2$–$Na_2(BO_2)_2$ as 6 : 1 : 3/4 : 9/2 = 24 : 4 : 3 : 18 or

$$14Ba_2Na_3(BO_2)_6F = 24Ba(BO_2)_2 + 4BaF_2 + 3Na_2F_2 + 18Na_2(BO_2)_2.$$

Note that the fractions of the salts in the compound that are present in the same ternary reciprocal system but with different masses in the complex vertices are related by the formula

$$C_iQ_i = c_iq_i, \tag{4}$$

where $C_i$, $Q_i$ are the fraction and mass of the $i$th salt in one system and $c_i$, $q_i$ are the same parameters in the other system.

For example, in the $Ba(BO_2)_2$–$BaF_2$–$NaF$–$NaBO_2$–$NaF$ system with masses in the complex vertices $q_{Ba(BO2)2} = 7$, $q_{BaF} = 3$, $q_{NaF} = 2$, and $q_{NaBO_2} = 4$, the salts in compound $D$ are related as 12 : 2 : 3 : 18, whereas in the 1/8 $Ba(BO_2)_2$–1/2 $BaF_2$–NaF–1/4 $NaBO_2$ system with the masses $m_{Ba(BO_2)_2} = 7/8$, $m_{BaF_2} = 3/2$, $m_{NaF} = 2$, and $m_{NaBO_2} = 1$, they are related as 96 : 4 : 3 : 72. If we introduce the designations $q_{Ba(BO_2)_2} = 7$, $q_{BaF} = 3$, $q_{NaF} = 2$, $q_{NaBO_2} = 4$, $c_{Ba(BO_2)_2} = 12$, $c_{BaF} = 2$, $c_{NaF} = 3$, and $c_{NaBO_2} = 18$ for the system $Ba(BO_2)_2$–$BaF_2$–NaF–$NaBO_2$ and $Q_{Ba(BO_2)_2} = 7/8$, $Q_{BaF_2} = 3/2$, $Q_{NaF} = 2$, $Q_{NaBO_2} = 1$, $C_{Ba(BO_2)_2} = 96$, $C_{BaF} = 4$, $C_{NaF} = 3$, and $C_{NaBO_2} = 72$, we obtain

$$C_{Ba(BO_2)_2}Q_{Ba(BO_2)_2} = 96 \cdot 7/8 = c_{Ba(BO_2)_2}q_{Ba(BO_2)_2} = 12 \cdot 7 = 84,$$

$$C_{BaF_2}Q_{BaF_2} = 4 \cdot 3/2 = c_{BaF_2}q_{BaF_2} = 2 \cdot 3 = 6,$$

$$C_{NaF}Q_{NaF} = 3 \cdot 2 = c_{NaF}q_{NaF} = 3 \cdot 2 = 6,$$

$$C_{NaBO_2}Q_{NaBO_2} = 72 \cdot 1 = c_{NaBO_2}q_{NaBO_2} = 18 \cdot 4 = 72.$$

Formula (4) makes it easier to determine the ratio of salts (i.e., their coefficients in the decomposition equation) in a system with specified masses in the complex vertices according to the known ratios of the same salts but with other masses in the vertices.

For example, the $Ba(BO_2)_2$–$BaF_2$–$Na_2F_2$–$Na_2(BO_2)_2$ system is indeed reciprocal because the following exchange reaction (without equating coefficients) occurs in it:

**Table 1.** Coordinates (2/24, 3/24, 6/24, 12/24, 1/24 = 0.083, 0.125, 0.250, 0.500, 0.042) of compound $D = Ba_2Na_3B_6O_{12}F$ of Ba–Na–B–O–F system in the subsystems

| Subsystem | Masses of material points $q_i$ | Component ratios | B coordinates | |
|---|---|---|---|---|
| | | | molar | mass |
| a) in $Ba(BO_2)_2$–$BaF_2$–$NaBO_2$ subsystem of $Ba(BO_2)_2$–$BaF_2$–NaF–$NaBO_2$ system | | | | |
| $1/7BaB_2O_4$–$1/3BaF_2$–$1/4NaBO_2$ | $p_i = 1$ | 7 : 1 : 8 | 7/16, 1/16, 8/16 = 0.44, 0.06, 0.50 | 0.54, 0.14, 0.32 |
| $1/7Ba(BO_2)_2$–$1/3BaF_2$–$1/2NaF$–$1/4NaBO_2$ | $p_i = 1$ | 14 : 1 : 1 : 12 | | |
| $Ba(BO_2)_2$–$BaF_2$–$NaBO_2$ | $q_{BaB_2O_4} = 7$, $q_{BaF_2} = 3$, $q_{NaBO_2} = 4$ | 3 : 1 : 6 | 3/10, 1/10, 6/10 = 0.3, 0.1, 0.6 | 0.54, 0.14, 0.32 |
| $Ba(BO_2)_2$–$BaF_2$–NaF–$NaBO_2$ | $q_{BaB_2O_4} = 7$, $q_{BaF_2} = 3$, $q_{NaF} = 2$, $q_{NaBO_2} = 4$ | 12 : 2 : 3 : 18 | | |
| $Ba(BO_2)_2$–$BaF_2$–$Na_2(BO_2)_2$ | $q_{BaB_2O_4} = 7$, $q_{BaF_2} = 3$, $q_{NaBO_2} = 4$ | 3 : 1 : 3 | 3/7, 1/7, 3/7 = 0.43, 0.14, 0.43 | 0.54, 0.14, 0.32 |
| $Ba(BO_2)_2$–$BaF_2$–$Na_2F_2$–$Na_2(BO_2)_2$ | $q_{BaB_2O_4} = 7$, $q_{BaF_2} = 3$, $q_{NaF} = 4$, $q_{NaBO_2} = 8$ | 24 : 4 : 3 : 18 | | |
| b) in BaO–$Na_2O$–$B_2O_3$–$BaF_2$ subsystem of BaO–$Na_2O$–$B_2O_3$–$BF_2$–NaF–$BF_3$ system | | | | |
| $1/2BaO$–$1/3Na_2O$–$1/5B_2O_3$–$BaF_2$ | $p_i = 1$ | 2 : 3 : 10 : 1 | 2/16, 3/16, 10/16, 1/16 = 0.12, 0.19, 0.63, 0.06 | 0.37, 0.15, 0.34, 0.14 |
| $1/2BaO$–$1/3Na_2O$–$1/5B_2O_3$–$1/3BF_2$–$1/2NaF$–$1/4BF_3$ | $p_i = 1$ | 16 : 18 : 60 : 1 : 1 : 4 | | |
| BaO–$Na_2O$–$B_2O_3$–$BaF_2$ | $q_{BaO} = 2$, $q_{Na_2O} = 3$, $q_{B_2O_3} = 5$, $q_{BaF_2} = 3$ | 3 : 3 : 6 : 1 | 3/13, 3/13, 6/13, 1/13 = 0.23, 0.23, 0.46, 0.08 | 0.37, 0.15, 0.34, 0.14 |
| BaO–$Na_2O$–$B_2O_3$–$BF_2$–NaF–$BF_3$ | $q_{BaO} = 2$, $q_{Na_2O} = 3$, $q_{B_2O_3} = 5$, $q_{BaF_2} = 3$, $q_{NaF} = 2$, $q_{BF_3} = 4$ | 48 : 36 : 72: 2 : 3 : 6 | | |
| BaO–$Na_2O$–$B_{2/3}O$–$BaF_2$ | $q_{BaO} = 2$, $q_{Na_2O} = 3$, $q_{B_2O_3} = 5/3$, $q_{BaF_2} = 3$ | 3 : 3 : 18 : 1 | 3/25, 3/25, 18/25, 1/25 = 0.12, 0.12, 0.72, 0.04 | 0.37, 0.15, 0.34, 0.14 |
| BaO–$Na_2O$–$B_{2/3}O$–$BF_2$–$Na_2F_2$–$B_{2/3}F_2$ | $q_{BaO} = 2$, $q_{Na_2O} = 3$, $q_{B_2O_3} = 5/3$, $q_{BaF_2} = 3$, $q_{NaF} = 4$, $q_{BF_3} = 8/3$ | 96 : 72 : 432 : 4 : 3 : 18 | | |

$$Ba(BO_2)_2 + Na_2F_2 = Na_2(BO_2)_2 + BaF_2.$$

The coefficients in the system $Ba(BO_2)_2$–$BaF_2$–NaF–$NaBO_2$ are chosen so as to equalize the contents of the corresponding cations and anions; therefore, exchange reactions also occur in this system as follows:

$$Ba(BO_2)_2 + 2NaF = 2NaBO_2 + BaF_2.$$

These two systems are identical because the exchange reactions in both involve a doubled number of sodium cations (with corresponding doubling of the number of anions).

At the same time, the $1/7\ Ba(BO_2)_2$–$1/3\ BaF_2$–$1/2\ NaF$–$1/4\ NaBO_2$ system at unit masses at the complex vertices ($p_i = 1$) cannot be called reciprocal because one must choose the equalizing coefficients (thus increasing the masses in the complex vertices):

$$7[1/7Ba(BO_2)_2] + 4[1/2NaF] = 8[1/4NaBO_2] + 3[1/3BaF_2].$$

Correspondingly, it is fairly difficult to determine the ratios $c_{BaB_2O_4} : c_{BaF_2} : c_{NaF} : c_{NaBO_2}$ for the salts while specifying compound $D$ because the coefficients 1/7 and 1/3 stand before the $Ba^+$ cation and the coefficients 2/7 and 1/2 are before the $BO_2^-$ anion. However, these ratios can be calculated from formula (4) using the relationship $C_{BaB_2O_4} : C_{BaF_2} : C_{NaF} : C_{NaBO_2} = 12 : 2 : 3 : 18$ in the $Ba(BO_2)_2$–$BaF_2$–NaF–$NaBO_2$ system with the masses $Q_{Ba(BO_2)_2} = 7$, $Qm_{BaF} = 3$, $Q_{NaF} = 2$, and $Q_{NaBO_2} = 4$ in the complex vertices:

$$c_{BaB_2O_4} = C_{BaB_2O_4} Q_{BaB_2O_4}/q_{BaB_2O_4} = 12 \cdot 7/1 = 12 \cdot 7 = 84,$$

$$c_{BaF_2} = C_{BaF_2} Q_{BaF_2}/q_{BaF_2} = 2 \cdot 3/1 = 6,$$

$$c_{NaF} = C_{NaF} Q_{NaF}/q_{NaF} = 3 \cdot 2/1 = 6,$$

$$c_{NaBO_2} = C_{NaBO_2} Q_{NaBO_2} / q_{NaBO_2} = 18 \cdot 4/1 = 72.$$

Now, we can write the following equation (with substitution of the obtained coefficients $c_i$:

$$7Ba_2Na_3B_6O_{12}F = 84(1/7BaB_2O_4) + 6(1/3BaF_2) + 6(1/2NaF) + 72(1/4NaBO_2).$$

Finally, in the $1/7Ba(BO_2)_2$–$1/3BaF_2$–$1/2NaF$–$1/4NaBO_2$ system, compound $D$ is set by the salt ratios 84 : 6 : 6 : 72 or 14 : 1 : 1 : 12.

The molar coordinates (7/16, 1/16, 8/16) of compound $D$ in the $1/7BaB_2O_4$–$1/3BaF_2$–$1/4NaBO_2$ system are as follows:

$$b_1 = \frac{(7/16)\cdot(1/7)M_{Ba(BO_2)_2}}{(7/16)\cdot(1/7)M_{Ba(BO_2)_2} + (1/16)\cdot(1/3)M_{BaF_2} + (8/16)\cdot(1/4)M_{NaBO_2}} = 0.54, \quad b_2 = 0.14, \quad b_3 = 0.32,$$

where $M_{Ba(BO_2)_2} = M_{Ba} + 2M_B + 4M_O = 137.34 + 2 \cdot 10.81 + 4 \cdot 15.99 = 190.96$; $M_{BaF_2} = M_{Ba} + 2M_F = 137.34 + 2 \cdot 18.99 = 171.34$; and $M_{NaBO_2} = M_{Na} + M_B + 2M_O = 22.99 + 10.81 + 2 \cdot 15.99 = 65.80$.

One can verify that, when passing to mass fractions, both the coordinates (3/10, 1/10, 6/10) in the $BaB_2O_4$–$BaF_2$–$NaBO_2$ system and the coordinates (3/7, 1/7, 3/7) in the $BaB_2O_4$–$BaF_2$–$Na_2(BO_2)_2$ system of compound $D$ will yield the same values, i.e., $b_1 = 0.540$, $b_2 = 0.142$, and $b_3 = 0.319$ (Table 1a).

Similarly, one can estimate the position of the point that corresponds to compound $D = Ba_2Na_3(B_3O_6)_2F$ in the $BaO$–$Na_2O$–$B_2O_3$–$BaF_2$ subsystem of the quaternary reciprocal system $BaO$–$Na_2O$–$B_2O_3$–$BaF_2$–$NaF$–$BF_3$ with different masses at its vertices (Table 1b).

Thus, when the trapezoid $BaB_2O_4$–$NaF$–$BaF_2$–$NaBO_2$ inside the initial pentatope Ba–Na–B–O–F (Fig. 1) and the corresponding square $1/7BaB_2O_4$–$NaF$–$BaF_2$–$NaBO_2$ are transformed into squares with other amounts of initial materials at the vertices, the coordinates of the point corresponding to the compound $D = Ba_2Na_3(B_3O_6)_2F$ change. Correspondingly, different ratios of initial salts are required to form it. For example, in the square $Ba(BO_2)_2$–$BaF_2$–$NaF$–$NaBO_2$, the salts $Ba(BO_2)_2$, BaF, and $NaBO_2$ must be taken in the ratio 3 : 1 : 6, whereas the salts $Ba(BO_2)_2$, NaF, and $NaBO_2$ must be taken in the ratio 2 : 1 : 2. In the other square with doubled masses of sodium salts, the salts $Ba(BO_2)_2$, $BaF_2$, and $Na_2(BO_2)_2$ are taken in the ratio 3 : 1 : 3, while the salts $Ba(BO_2)_2$, $(NaF)_2$, and $(NaBO_2)_2$ are taken in the ratio 4 : 1 : 2. These two pairs of versions of expressions for concentrations correspond to the accepted molar and equivalent ways of setting the amounts of the initial materials. When similar multidimensional figures are polyhedrated, it is important to trace the masses of material points at the vertices of polyhedra of the same or smaller dimensions in the systems described by different authors in different sources and using different ways to express the concentration.

## POLYHEDRATION

*The main purposes of polyhedration* of the concentration space of a multicomponent system are as follows:

(i) to describe the boundaries of all subsystems of multidimensional concentration complex;

(ii) to establish relationships between the local systems of B coordinates of subsystems in the unified concentration space of multicomponent system;

(iii) to determine the affiliation of an arbitrarily specified composition of a multidimensional complex of concentration subsystem.

*To solve the first problem*, we developed a program for analyzing the results of a partition of a multicomponent system by quasi-binary cross sections and (hyper)planes generated by them into subsystems. Depending on the number of compounds formed and/or the dimension of the concentration space, this problem may be so cumbersome that it is not expedient or even impossible to solve it without a computer.

To solve the *second* and *third* (which is inverse to second) *problems* in the determination of each $i$th subsystem, the linking equation of type (1) can be written multiply in the form $Z = K_iX_i$, and the condition for the affiliation of composition $G$ to the subsystem $X(x_i)$ is the inequality

$$0 \le x_i \le 1$$

for all of its $m$ normalized coordinates $x_i$.

**Kraeva's polyhedration algorithm.** Most often, polyhedration is performed by the method based on the interpretation of triangulated concentration space in the form of a graph [13]. Polyhedra are recognized using an algorithm that was used in the theory of graphs when constructing a set of independent subsets based on the adjacency matrix and its list [14]. Among the two possible ways of describing a graph using an adjacency list (containing unit elements of the adjacency matrix) or nonadjacency list (containing zero matrix elements), the second technique is generally preferred.

The essence of this algorithm is as follows:

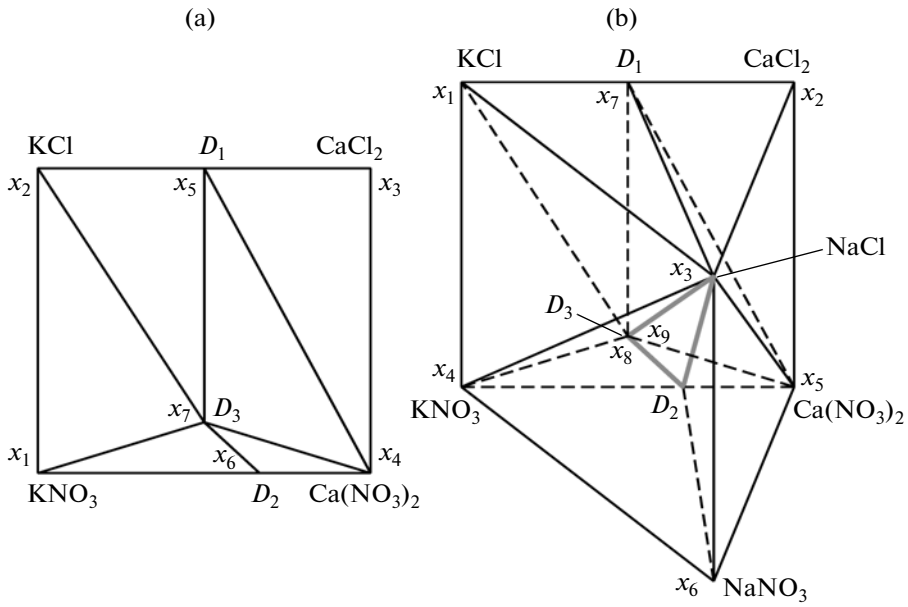


**Fig. 2.** Polyhedration of (a) ternary reciprocal system K, Ca || $NO_3$, Cl and (b) quaternary reciprocal system K, Ca, Na || $NO_3$, Cl with binary compounds $D_1 = KCl \cdot CaCl_2$ and $D_2 = KNO_3 \cdot 2Ca(NO_3)_2$ and quasi-quaternary compound $K_2Ca_2(NO_3)_5Cl$ [16] or $D_3 = 5KNO_3 \cdot KCl \cdot CaCl_2 \cdot 5Ca(NO_3)_2$.

(i) to enumerate ($x_i$) and present the initial components and stoichiometric compounds in the form of a graph;

(ii) to write the triangular adjacency matrix $R$, in which $r_{i,j} = 1$ if the element $x_i$ is connected with $x_j$ and $r_{i,j} = 0$ if these elements are not connected (because, if there is only one connection between two vertices and the graph is unoriented, one can consider the adjacency matrix as triangular, having denoted the diagonal elements as $r_{i,i} = *$);

(iii) to compile (based on the adjacency matrix) the adjacency list $R_L$, in which vertices nonadjacent to $x_i$ are written in each $i$th row $C_i$;

(iv) to multiply the rows of the adjacency list $C_i$ with allowance for the absorption rule in order to obtain the sum $\Sigma S_j$, $j = \overline{1, f}$, where $f$ is the number of simplices into which a given polyhedron can be divided;

(v) to perform inversion, i.e., to rewrite all $S_i$ terms so as to include the absent graph vertices.

For example, to perform the triangulation of the K,Ca||$NO_3$,Cl system (Fig. 2a), numbers are assigned to the vertices of the square $KNO_3 = x_1$, $KCl = x_2$, $CaCl_2 = x_3$, $Ca(NO_3)_2 = x_4$ and to the compounds $D_1 = KCl \cdot CaCl_2$, $= x_5$, $D_2 = KNO_3 \cdot 2Ca(NO_3)_2 = x_6$, and $D_3 = KNO_3 \cdot 2Ca(NO_3)_2 \cdot KCl = x_7$, after which the adjacency matrix and adjacency list are written as follows:

$$R = \begin{array}{c} \\ x_1 \\ x_2 \\ x_3 \\ x_4 \\ x_5 \\ x_6 \\ x_7 \end{array} \begin{array}{c} \begin{array}{ccccccc} x_1 & x_2 & x_3 & x_4 & x_5 & x_6 & x_7 \end{array} \\ \left\| \begin{array}{ccccccc} * & 1 & 0 & 0 & 0 & 1 & 1 \\ & * & 0 & 0 & 1 & 0 & 1 \\ & & * & 1 & 1 & 0 & 0 \\ & & & * & 1 & 1 & 1 \\ & & & & * & 0 & 1 \\ & & & & & * & 1 \\ & & & & & & * \end{array} \right\| \end{array},$$

$$R_L = \begin{array}{c} \\ x_1 \\ x_2 \\ x_3 \\ x_5 \end{array} \begin{array}{c} \begin{array}{ccc} & & \end{array} \\ \left\| \begin{array}{ccc} x_3 & x_4 & x_5 \\ x_3 & x_4 & x_6 \\ x_6 & x_7 & \\ x_6 & & \end{array} \right\| \end{array}.$$

Then, the rows of the adjacency list are successively multiplied as follows:

$$(x_1 + x_3x_4x_5)(x_2 + x_3x_4x_6) = x_1x_2 + x_1x_3x_4x_6 + x_2x_3x_4x_5 + x_3x_4x_5x_6.$$

Multiplication is performed according to the absorption rule: when multiplying by the third row $(x_3 + x_6x_7)$,

$$(x_1x_2 + x_1x_3x_4x_6 + x_2x_3x_4x_5 + x_3x_4x_5x_6)(x_3 + x_6x_7) = x_1x_2x_3 + x_1x_3x_4x_6 + x_2x_3x_4x_5 + x_3x_4x_5x_6 + x_1x_2x_6x_7 + x_1x_3x_4x_6x_7 + x_2x_3x_4x_5x_6x_7 + x_3x_4x_5x_6x_7,$$

the term $x_1x_3x_4x_6x_7$ is cancelled out because it contains another term: $x_1x_3x_4x_6$. Similarly, we exclude two more terms: $x_2x_3x_4x_5x_6x_7$ and $x_3x_4x_5x_6x_7$, which contain $x_2x_3x_4x_5$ and $x_3x_4x_5x_6$, respectively. As a result, after

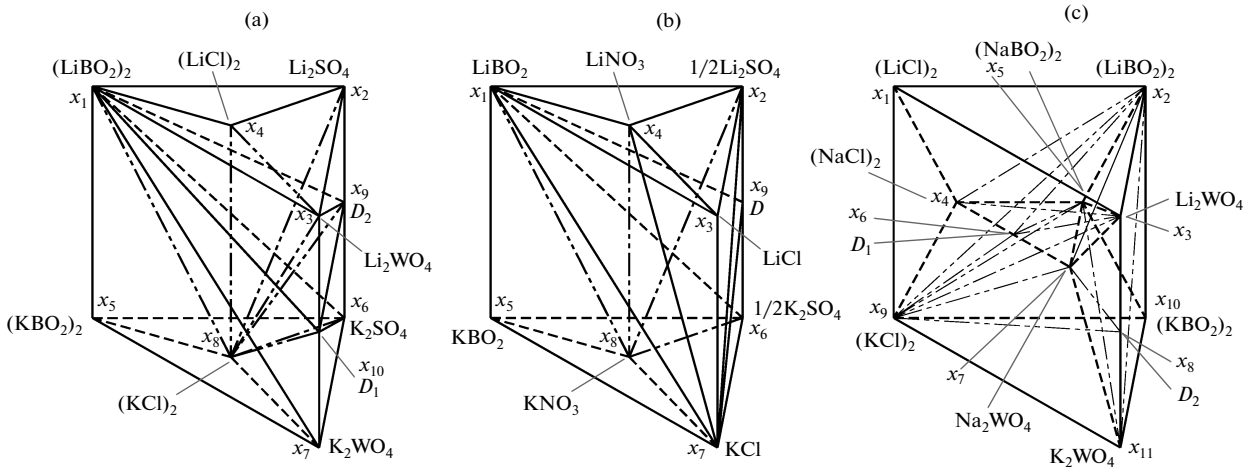


**Fig. 3.** Polyhedration of quinary reciprocal systems (a) Li, K || $BO_2$, $SO_4$, $WO_4$, Cl, (b) Li, K || $BO_2$, $SO_4$, Cl, $NO_3$, and (c) Li, Na, K || Cl, $BO_2$, $WO_4$.

multiplication by the third row of the adjacency list, the following sum remains:

$$x_1x_2x_3 + x_1x_3x_4x_6 + x_2x_3x_4x_5 + x_3x_4x_5x_6 + x_1x_2x_6x_7,$$

which is then multiplied by the fourth row of the adjacency list:

$$\begin{aligned}(x_1x_2x_3 + x_1x_3x_4x_6 + x_2x_3x_4x_5 + x_3x_4x_5x_6\\ + x_1x_2x_6x_7)(x_5 + x_6) = x_1x_2x_3x_5 + x_1x_3x_4x_5x_6\\ + x_2x_3x_4x_5 + x_3x_4x_5x_6 + x_1x_2x_5x_6x_7 + x_1x_2x_3x_6\\ + x_1x_3x_4x_6 + x_2x_3x_4x_5x_6 + x_3x_4x_5x_6 + x_1x_2x_6x_7.\end{aligned}$$

We cancel the terms of this sum, which contain its other terms : $x_1x_2x_5x_6x_7$ and $x_2x_3x_4x_5x_6$, which contain, respectively, $x_1x_2x_6x_7$ and $x_2x_3x_4x_5$, as well as the product $x_1x_3x_4x_5x_6$, which includes two terms: $x_1x_3x_4x_6$ and $x_3x_4x_5x_6$.

As a result, polyhedration performed based on the adjacency list is reduced to the multiplication of its rows in the form

$$\begin{aligned}(x_1 + x_3x_4x_5)(x_2 + x_3x_4x_6)(x_3 + x_6x_7)(x_5 + x_6)\\ = x_1x_2x_3x_5 + x_2x_3x_4x_5 + x_3x_4x_5x_6 + x_1x_2x_3x_6\\ + x_1x_3x_4x_6 + x_1x_2x_6x_7.\end{aligned}$$

The thus obtained terms cannot be physicochemically or geometrically interpreted. However, when inverting them (i.e., replacing each term by a similar product with absent polyhedron vertices), one obtains a list of simplices into which the polyhedron is divided. For example, the term $x_1x_2x_3x_5$ is rewritten in the form $x_4x_6x_7$, etc. After the inversion, the vertices of six simplices (triangles) are enumerated:

$$x_4x_6x_7 + x_1x_6x_7 + x_1x_2x_7 + x_4x_5x_7 + x_2x_5x_7 + x_3x_4x_5.$$

These are the simplices into which the square of the K,Ca||$NO_3$,Cl system is divided. The polyhedration of the quaternary reciprocal system K,Ca,Na||$NO_3$,Cl (Fig. 2b) is performed similarly.

Polyhedration of the Li,K||$BO_2$,$SO_4$,$WO_4$,Cl system with compounds $D_1 = Li_2WO_4 \cdot K_2WO_4$ and $D_2 = Li_2SO_4 \cdot K_2SO_4$ is completed by six pentatopes separated by five tetrahedra (Fig. 3a), while the polyhedration of the Li,K||$BO_2$,$SO_4$,Cl,$NO_3$ system with compound $D = Li_2SO_4 \cdot K_2SO_4$ is completed by five pentatopes with four cutting tetrahedra (Fig. 3b).

After the polyhedration of the Li, Na, K||Cl, $BO_2$, $WO_4$ system with compounds $D_1 = Na_2WO_4 \cdot (NaCl)_2$ and $D_2 = Li_2WO_4 \cdot K_2WO_4$, we have nine pentatopes and ten cutting tetrahedra (Fig. 2c).

**Limitations of the polyhedration algorithm in the presence of internal points.** According to Bergman's classification [15], a binary compound of a ternary reciprocal system belongs to the diagonal of the composition square, a ternary compound is located in the triangle separated by the stable diagonal, and a quaternary compound (in our opinion, it is more convenient to call it quasi-quaternary) is formed from all four initial salts.

For example, in the K,Ca||$NO_3$,Cl system, the stoichiometry $KNO_3 \cdot 2Ca(NO_3)_2 \cdot KCl$ is assigned to the "ternary hetero-compound" $D_3 = K_2Ca_2(NO_3)_5Cl$ [16], although it can also be formally written as $4KNO_3 \cdot CaCl_2 \cdot 3Ca(NO_3)_2$. At the same time, the absence of stable diagonal in the composition square and the connection of this compound with three out of the four vertices of the square and with compounds $D_1 = KCl \cdot CaCl_2$ and $D_2 = KNO_3 \cdot 2Ca(NO_3)_2$ (Fig. 2a) give us grounds to consider it as a quasi-quaternary compound $5KNO_3 \cdot KCl \cdot CaCl_2 \cdot 5Ca(NO_3)_2$.

Similarly, proceeding from Bergman's terminology, if there were no stable diagonals in the Ba,Na||F,$BO_2$ system, the $Ba_2Na_3(B_3O_6)_2F$ compound should be considered quasi-quaternary. In this paper, the $BaF_2$–$NaBO_2$ diagonal is considered to be stable, and compound $D$ is considered to be ternary and to belong to the $Ba(BO_2)_2$–$BaF_2$–$NaBO_2$ subsystem.

When binary and quasi-quaternary compounds are formed, there are no problems with triangulation. For example, in the case of the triangulation of the

**Table 2.** Results of polyhedration of quinary reciprocal systems Li,K||$BO_2$,$SO_4$,$WO_4$,Cl, Li,K||$BO_2$,$SO_4$,Cl,$NO_3$ and Li,Na,K||Cl,$BO_2$,$WO_4$ (Fig. 3)

| System | Adjacency list $R_L$ | Simplices |
|---|---|---|
| Li,K||$BO_2$,$SO_4$,$WO_4$,Cl<br>$D_1 = Li_2WO_4 \cdot K_2WO_4$<br>$D_2 = Li_2SO_4 \cdot K_2SO_4$ | $x_2 \parallel x_5\ x_6\ x_7\ x_{10}$<br>$x_3 \parallel x_5\ x_6\ x_7$<br>$x_4 \parallel x_5\ x_6\ x_7\ x_9\ x_{10}$<br>$x_5 \parallel x_9\ x_{10}$<br>$x_7 \parallel x_9$ | $x_1x_6x_8x_9x_{10} = (LiBO_2)_2{-}K_2SO_4{-}(KCl)_2{-}D_1{-}D_2$<br>$x_1x_3x_8x_9x_{10} = (LiBO_2)_2{-}Li_2WO_4{-}(KCl)_2{-}D_1{-}D_2$<br>$x_1x_2x_3x_8x_9 = (LiBO_2)_2{-}Li_2SO_4{-}Li_2WO_4{-}(KCl)_2{-}D_1$<br>$x_1x_2x_3x_4x_8 = (LiBO_2)_2{-}Li_2SO_4{-}Li_2WO_4{-}(LiCl)_2{-}(KCl)_2$<br>$x_1x_6x_7x_8x_{10} = (LiBO_2)_2{-}K_2SO_4{-}K_2WO_4{-}(KCl)_2{-}D_2$<br>$x_1x_5x_6x_7x_8 = (LiBO_2)_2{-}(KBO_2)_2{-}K_2SO_4{-}K_2WO_4{-}(KCl)_2$ |
| Li,K||$BO_2$,$SO_4$,Cl,$NO_3$<br>$D = Li_2SO_4 \cdot K_2SO_4$ | $x_2 \parallel x_5\ x_6$<br>$x_3 \parallel x_5\ x_6\ x_8\ x_9$<br>$x_4 \parallel x_5\ x_6\ x_9$<br>$x_5 \parallel x_9$ | $x_1x_6x_7x_8x_9 = LiBO_2{-}1/2K_2SO_4{-}KCl{-}KNO_3{-}D$<br>$x_1x_2x_7x_8x_9 = LiBO_2{-}1/2Li_2SO_4{-}KCl{-}KNO_3{-}D$<br>$x_1x_2x_4x_7x_8 = LiBO_2{-}1/2Li_2SO_4{-}LiNO_3{-}KCl{-}KNO_3$<br>$x_1x_2x_3x_4x_7 = LiBO_2{-}1/2Li_2SO_4{-}LiCl{-}LiNO_3{-}KCl$<br>$x_1x_5x_6x_7x_8 = LiBO_2{-}KBO_2{-}1/2K_2SO_4{-}KCl{-}KNO_3$ |
| Li,Na,K||Cl,$BO_2$,$WO_4$<br>$D_1 = Na_2WO_4 \cdot (NaCl)_2$<br>$D_2 = Li_2WO_4 \cdot K_2WO_4$ | $x_1 \parallel x_5\ x_6\ x_7\ x_8\ x_{10}\ x_{11}$<br>$x_3 \parallel x_5\ x_{10}\ x_{11}$<br>$x_4 \parallel x_7\ x_8\ x_{10}\ x_{11}$<br>$x_5 \parallel x_8$<br>$x_6 \parallel x_8\ x_{10}\ x_{11}$<br>$x_7 \parallel x_{10}$<br>$x_8 \parallel x_{10}$ | $x_2x_7x_8x_9x_{11} = (LiBO_2)_2{-}Na_2WO_4{-}D_2{-}(KCl)_2{-}K_2WO_4$<br>$x_2x_5x_9x_{10}x_{11} = (LiBO_2)_2{-}(NaBO_2)_2{-}(KCl)_2{-}(KBO_2)_2{-}K_2WO_4$<br>$x_2x_5x_7x_9x_{11} = (LiBO_2)_2{-}(NaBO_2)_2{-}Na_2WO_4{-}(KCl)_2{-}K_2WO_4$<br>$x_2x_5x_6x_7x_9 = (LiBO_2)_2{-}(NaBO_2)_2{-}D_1{-}Na_2WO_4{-}(KCl)_2$<br>$x_2x_4x_5x_6x_9 = (LiBO_2)_2{-}(NaCl)_2{-}(NaBO_2)_2{-}D_1{-}(KCl)_2$<br>$x_2x_3x_7x_8x_9 = (LiBO_2)_2{-}Li_2WO_4{-}Na_2WO_4{-}D_2{-}(KCl)_2$<br>$x_2x_3x_6x_7x_9 = (LiBO_2)_2{-}Li_2WO_4{-}D_1{-}Na_2WO_4{-}(KCl)_2$<br>$x_2x_3x_4x_6x_9 = (LiBO_2)_2{-}Li_2WO_4{-}(NaCl)_2{-}D_1{-}(KCl)_2$<br>$x_1x_2x_3x_4x_9 = (LiCl)_2{-}(LiBO_2)_2{-}Li_2WO_4{-}(NaCl)_2{-}(KCl)_2$ |

K,Ca||$NO_3$,Cl system with the quasi-quaternary compound $5KNO_3 \cdot KCl \cdot CaCl_2 \cdot 5Ca(NO_3)_2$, Kraeva's algorithm allows one to recognize all six simplices (Fig. 2a).

However, this algorithm cannot not recognize the "internal" point of the ternary compound, which is located in the corresponding simplex. For example, in the case of the triangulation of the Ba,Na||F,$BO_2$ system based on the assumption of the stability of the $BaF_2$–$NaBO_2$ diagonal with ternary compound $D$ = $Ba_2Na_3(B_3O_6)_2F$, one can only see the $BaF_2$–NaF–$NaBO_2$ simplex and $Ba(BO_2)_2$–$BF_2$–$NaBO_2$–$D$ microcomplex with the internal point $D$ (Fig. 1).

We have a similar situation with the ternary compound $Na_2Sr(VO_3)_4$ in the $Na_2O$–$V_2O_5$–SrO system; this compound is related via quasi-binary cross sections with the binary compounds $NaVO_3$, $Na_{2,5}V_6O_{16}$ and $Sr_2V_2O_7$ (p. 99 in [3]). The aforementioned algorithm distinguishes only the $NaVO_3$–$Na_{2.5}V_6O_{16}$–$Sr_2V_2O_7$–$Na_2Sr(VO_3)_4$ microcomplex.

Thus, when complexes with internal points are polyhedrated, Kraeva's algorithm has certain limitations. To remove these points, these internal vertices can be excluded at the first stage, and primary triangulation can be performed without them. Then, it is necessary to find out to which obtained simplices the remaining internal points belong and to perform an additional partition of the formed microcomplexes. In other words, when performing triangulation of the Ba,Na||F,$BO_2$ system, one must first perform triangulation without compound $D$; then, if $D \in \{Ba(BO_2)_2$–$BF_2$–$NaBO_2\}$, we connect $D$ with the vertices $Ba(BO_2)_2$, $BF_2$, and $NaBO_2$.

## POLYHEDRATION TESTING

Manual polyhedration [13] leads to cumbersome calculations and is unreasonable without corresponding software. However, when using a program, one must verify both the correctness of the introduction of the initial information into the adjacency matrix and the results obtained at the output in a particular way. Therefore, to verify elements of the adjacency list introduced into the program, we derived relations that linked the number of adjacency matrix elements (graph vertices and bonds between them) with the number of internal diagonal planes and simplices and made it possible to estimate a priori the result of simplexing.

**Triangulation testing.** The triangulation rules (in the condensed form) for ternary systems with intermediate phases of constant composition reflect the

known analytical relations (see, e.g., [17]) between the triangulation parameters:

$$R = M + 3S, \quad T = 1 + M + 2S,$$

where $R$ is the number of quasi- and partially quasi-binary cuts, $T$ is the number of secondary systems, and $M$ and $S$ are the numbers of binary and ternary compounds. At $S = 0$, these relations are simplified to $R = M$ and $T = 1 + M$.

The application of the Euler equation to planar $n$-vertex polygons with $m$ internal points $(n + m) - (n + d) + f = 1$ led to expressions that relate the numbers of polygon vertices ($n$) and internal points ($m$) with the numbers of diagonals ($d$) and simplices (triangles) ($f$) as follows [18]:

$$d = 3m + n - 3, \quad f = 2m + n - 2.$$

Having selected the number of points on the polygon sides ($k$), one arrives at the expressions

$$d = n + k + 3m - 3, \quad f = n + k + 2m - 2. \qquad (5)$$

The total number $t$ of all points (graph vertices), the number $d$ of internal diagonals, and the number $f$ of simplices (triangles) obtained as a result of triangulation are related by the Euler equation

$$(n + k + m) - (r + d) + f = 1 \quad \text{or} \quad t - p + f = 1, \qquad (6)$$

where $r$ is the number of segments on the sides of the triangle into which they are divided by $k$ points.

At $n = 3$, for the concentration triangle ($r = 3 + k$), expressions (5) and (6) take the form

$$\begin{aligned} (3 + k + m) - (r + d) + f &= 1, \\ d = k + 3m, \quad f &= 1 + k + 2m. \end{aligned} \qquad (7)$$

At $n = 4$, the topological relations (5) and (6) for simplexing concentration complexes of ternary reciprocal systems can be written as

$$\begin{aligned} (4 + k + m) - (r + d) + f &= 1, \\ d = 1 + k + 3m, \quad f &= 2 + k + 2m. \end{aligned} \qquad (8)$$

Thus, expression (6) relates the number $t$ of rows (columns) of the adjacency matrix and number $p$ of its unit elements with the numbers of one-dimensional ($d$) and two-dimensional ($f$) simplices.

Note that the number of sides and diagonals of the complex $p = r + d$ is equal to the number of unit elements in the adjacency matrix. Since $t \cdot t$ is the number of all elements of square adjacency matrix and $(t \cdot t - t)/2$ is the number of all elements of its upper half, the number $p$ can be calculated from the formula

$$p = \frac{t \cdot t - t}{2} - t_0, \qquad (9)$$

in which $t_0$ is the number of adjacency list elements (or the number of zero elements of the adjacency matrix).

**Tetrahedration testing.** For quaternary systems divided into tetrahedra, the initial information is set by the sum $t = n + k + m + b$, where $n$ is the number of vertices of the initial concentration complex, $k$ is the number of points on the edges, $m$ is the number of points on the faces, and $b$ is the number of quaternary internal points, and by the number $p = r + d + q$, where $r$ is the number of edges (or their fragments), $d$ is the number of diagonals on the faces, and $q$ is the number of internal diagonals.

If we introduce the designation $h = f + g$, where $f$ is the number of external faces (or their fragments) and $g$ is the number of internal secant planes (simplices) and denote the number of three-dimensional simplices (tetrahedra) as $s$, the numbers of vertices ($t$), edges ($p$), faces ($h$), and simplices ($s$) will be related by a formula similar to (6):

$$t - p + h - s = 1 \qquad (10)$$

$$\text{or} \quad (n + k + m + b) - (r + d + q) + (f + g) - s = 1.$$

Each tetrahedron has four faces, each of which is taken into account twice if it is an internal diagonal plane and once if it coincides with an external face of the polyhedron. Therefore,

$$4s = f + 2g. \qquad (11)$$

The joint solution of (10) and (11) with respect to $g$ and $s$ yields formulas that make it possible to determine the number of internal triangles dividing the polyhedron into three-dimensional simplices (tetrahedra):

$$g = 2 - 2t + 2p - 3f/2, \quad s = 1 - t + p - f/2. \qquad (12)$$

Since $t$ is the total number of vertices of the initial concentration complex (i.e., binary, ternary, and quaternary compounds) and $p$ is the number of links between them, which is given by formula (9), the only thing to do is to estimate $f$ (the number of simplices on external faces).

The $ABCD$ tetrahedron ($n = 4$) has six edges and four triangular faces. When polyhedration is performed, the initial information in the adjacency matrix is set by the number $t$ of points and number $p$ of segments that connect them as follows: $t = 6 + k + m + b$ and $p = r + d + q$, where $k$ is the number points on edges, $m$ is the number of points on faces, $b$ is the number of points in the polyhedron, $r$ is the number of edges (or their segments), $d$ is the number of diagonals on faces, and $q$ is the number of internal diagonals. Edges are divided by binary compounds into $p$ fragments as follows: $r = 6 + k$.

The number $q$ of internal diagonals is expressed in terms of $p$, $k$, and $m$ as

$$\begin{aligned} q = p - r - d &= p - (6 + k) - (2k + 3m) \\ &= p - 6 - 3k - 3m. \end{aligned} \qquad (13)$$

Since the tetrahedron faces are ternary systems, the numbers of internal diagonals ($d$) and simplices ($f$) for them can be calculated using formulas (7):

$$\begin{aligned} d = (k_{AB} + k_{AC} + k_{BC} + 3m_{ABC}) \\ + (k_{AB} + k_{AD} + k_{BD} + 3m_{ABD}) + (k_{AC} + k_{AD} + k_{CD} \\ + 3m_{ACD}) + (k_{BC} + k_{BD} + k_{CD} + 3m_{BCD}), \end{aligned}$$

and

$$\begin{aligned} f = (1 + k_{AB} + k_{AC} + k_{BC} + 2m_{ABC}) + (1 + k_{AB} + k_{AD} \\ + k_{BD} + 2m_{ABD}) + (1 + k_{AC} + k_{AD} + k_{CD} + 2m_{ACD}) \\ + (1 + k_{BC} + k_{BD} + k_{CD} + 2m_{BCD}) \end{aligned}$$

or

$$d = 2k + 3m, \quad f = 4 + 2k + 2m. \tag{14}$$

In this case, expressions (12) are transformed to

$$\begin{aligned} g &= 2 - 2t + 2p - 3f/2 = 2 - 2(4 + k + m + b) \\ &+ 2(r + d + q) - 3(4 + 2k + 2m)/2 \\ &= k + m + 2q - 2b, \end{aligned} \tag{15}$$

$$\begin{aligned} s &= 1 - t + p - f/2 = 1 - (4 + k + m + b) \\ &+ (r + d + q) - (4 + 2k + 2m)/2 \\ &= 1 + k + m + q - b. \end{aligned} \tag{16}$$

Thus, to determine in advance the number $s$ of simplices (tetrahedra) and the number $g$ of internal cut planes from formulas (15) and (16), it is sufficient to have initial information about the numbers of binary, ternary, and quaternary compounds ($k$, $m$, and $b$, respectively) and the number $q$ of internal cut diagonals.

Since the $A, B \parallel X, Y, Z$ polyhedron has six vertices ($n = 6$), nine edges, two triangular faces, and three tetragonal faces, the edges are divided by $k$ points into $r$ segments: $r = 9 + k$, and the total number of points is $t = 6 + k + m + b$. Based on (7) and (8), one can calculate the numbers of diagonals ($d$) and two-dimensional simplices ($f$) on all triangular and square faces of the polyhedron:

$$\begin{aligned} d &= (k_{12} + k_{13} + k_{23} + 3m_{123}) \\ &+ (k_{45} + k_{46} + k_{56} + 3m_{456}) \\ &+ (1 + k_{12} + k_{14} + k_{25} + k_{45} + 3m_{1254}) \\ &+ (1 + k_{13} + k_{14} + k_{36} + k_{46} + 3m_{1364}) \\ &+ (1 + k_{23} + k_{25} + k_{36} + k_{56} + 3m_{2365}), \\ f &= (1 + k_{12} + k_{13} + k_{23} + 2m_{123}) \\ &+ (1 + k_{45} + k_{46} + k_{56} + 2m_{456}) \\ &+ (2 + k_{12} + k_{14} + k_{25} + k_{45} + 2m_{1254}) \\ &+ (2 + k_{13} + k_{14} + k_{36} + k_{46} + 2m_{1364}) \\ &+ (2 + k_{23} + k_{25} + k_{36} + k_{56} + 2m_{2365}); \end{aligned}$$

hence,

$$d = 3 + 2k + 3m, \quad f = 8 + 2k + 2m.$$

The number $q$ of internal diagonals is expressed in terms of $p$, $k$, and $m$ as follows:

$$\begin{aligned} q &= p - r - d = p - (9 + k) - (3 + 2k + 3m) \\ &= p - 12 - 3k - 3m. \end{aligned}$$

The numbers of internal planes ($g$) and tetrahedra ($s$) are expressed in terms of the initial numbers of points on the edges ($k$), on the faces ($m$), and inside the polyhedron ($b$) and the number $q$ of internal diagonals as follows:

$$g = 2 - 2t + 2p - 3f/2 = 2 + k + m + 2q - 2b,$$
$$s = 1 - t + p - f/2 = 3 + k + m + q - b.$$

## CONCLUSIONS

To make the polyhedration of the concentration space of multicomponent systems an automatic procedure, we developed a program that uses the presentation of divided multidimensional space in the form of a graph. This program is based on Kraeva's algorithm, in which the connections between the vertices of simplices and the initial complex are described in terms of the adjacency matrix. To use this program, one must enumerate all vertices of the complex, including those of the initial concentration polyhedron, as well as the binary, ternary, and more complex compounds, and introduce indices of zero elements of the adjacency matrix that correspond to unconnected vertices.

The program developed forms the adjacency matrix and adjacency list, compiles and calculates the product of adjacency list elements with allowance for the absorption rule, performs inversion, and yields a list of simplices with their enumerated vertices.

A limitation of Kraeva's method was revealed for the polyhedration of concentration complexes with internal vertices. To remove this limit, one can exclude these internal points in the first triangulation stage and perform primary triangulation without them. The next task is to determine which of the obtained simplices the remaining internal points belong to and perform an additional partition of the microcomplexes formed.

Geometrically, an $n$-dimensional complex (a physicochemical system), the vertices of which are the initial-graph vertices, is divided into fragments (subsystems). The algorithm forms families of subsets, each term of which can combine different numbers of vertices. Most often an $n$-dimensional complex is divided into $n$-dimensional simplices. However, in some cases, polyhedration may yield either simplices with smaller dimensions or complexes that require additional polyhedration.

The occurrence of $(n - 1)$-dimensional simplices or $(n + 1)$-dimensional complexes can be caused by an error in the initial data in the adjacency matrix or by deliberate removal or doubling of some connections when it is desirable to emphasize the presence of solid solutions.

If polyhedration yields a microcomplex, it is necessary to check whether one of its vertices belongs to some simplex from the four remaining vertices. If yes, the central point must be connected with the remaining vertices to obtain three-dimensional simplices. If no, either the microcomplex corresponds to a subsystem with a continuous series of solid solutions (the additional polyhedration is not required in this case) or an extra diagonal was erroneously introduced.

When it was necessary to perform additional polyhedration (in the cases where the maximum independent subset of graph was a complex), the main algorithm was supplemented with additional procedures, which exclude some graph vertices (and the connections of other vertices with them) in the first stage of polyhedration and then take into account these vertices in the second stage.

To verify the initial data and the results of polyhedration, we derived formulas linking the number of

vertices and edges of graph with the number of simplices and their boundaries.

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