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Tetracomponent System State Diagrams for Analysis of Material Compositions

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Chemical compounds, composites and raw material mixture compositions expressed by system state diagrams were analysed in this work. The amounts of components % are given, also other parameters (such as temperature, pressure, etc.) can be included. Generally monocomponent (one material, temperature and pressure coordinates), bicomponent (two materials, temperature coordinates) and tetracomponent (three materials, also isotherms can be shown) are used. In this paper, we examined the tetracomponent system state diagram formation, compound or composite composition inclusion into diagrams and also composition identification possibilities. It is shown that the composition can be introduced into the diagram also by the method which could be of interest to patent authors, because some of the information in the diagrams can acquire a "know–how" function.

Keywords: state diagrams, materials, compositions, compounds, composites.

1. INTRODUCTION

The graphical expression of interdependent connections between equilibrium physico-chemical systems state parameters (composition), also between them and other system parameters (concentrations, volume, pressure, thermodynamic parameters) is called the system's (materials, phase) state diagram.

State diagrams are very important in material research techniques, industry and mineralogy. For example, iron – carbon state diagrams are the bases for steel thermal processing. Metal – sulphur state diagrams are used very widely in non-ferrous metal metallurgy. Water – salt state diagrams are very important in metallurgy and other mineral salt technologies. Silicate state diagrams contain the information, without which it is not possible to investigate silicates (cements, foam cement concretes, ceramic materials, glasses, slags, composites).

Composite material state diagrams have a very important significance in production and research.

The state diagrams reflect the characteristics of multicomponent systems.

Monocomponent state diagrams contain information about water, metal, carbon, sulphur, silicon dioxide and other important material composition structure in techniques as well as property dependency on temperature and pressure [1 – 3].

Bicomponent state diagrams reflect analogical information about alloys (Fe-C, Cu-Zn, Ag-Cu, Sn-Sb, etc.), construction material basis – silicates, aluminates, etc., also about the materials themselves [4, 5].

Tetracomponent state diagrams provides us information and data about metal alloys, construction materials, composites and many other important material compositions. For example, the composition of sand, clay, tripoli, glauconite are expressed by $SiO_2 – (Al_2O_3 + Fe_2O_3) – (CaO + MgO)$ diagrams [6 – 8], limestone and opoka compositions – $CaCO_3 – SiO_2 – (Al_2O_3 + Fe_2O_3)$ [9 – 11], dolomite compositions – $(CaCO_3 + MgO) – SiO_2 – CO_2$ diagrams, Portland cement binder system $CaO – R_2O_3 – SiO_2$ [12], anhydrite cement – organic fillers – polymeric binders [13, 14], Portland cement – sand – foaming agent (surfactants) [15], aluminate cement – ferrochromium slag – microsilica ($CaO – Al_2O_3 – SiO_2$) [16], gypsum – organic fillers – polymeric binders [17, 18], etc.

Further on, we will discuss the tetracomponent state diagrams formation, component composition introduction into it and retrival of system composition information present in diagrams [19].

A major objective of the present research is to demonstrate that tetracomponent system state diagrams can be used to investigate and analyze compositions of many important and practically useful materials and systems, such as:

- metal alloys,
- sand, clay: $SiO_2 – (CaO + MgO) – (Al_2O_3 + Fe_2O_3)$ – ignition losses (practically CO_2 and H_2O),
- limestone, lime: $(CaO + MgO) – SiO_2 – (Al_2O_3 + Fe_2O_3)$ – ignition losses,
- tripoli, opoka: $SiO_2 – (Al_2O_3 + Fe_2O_3) – (CaO + MgO) – (Na_2O + K_2O)$,
- dolomite: $CaO – MgO – SiO_2$ – ignition losses,
- glauconite: $SiO_2 – (Al_2O_3 + Fe_2O_3) – (CaO + MgO) – (Na_2O + H_2O)$,
- Portland cement: $(CaO + MgO) – SiO_2 – (Al_2O_3 + Fe_2O_3) – (Na_2O + K_2O)$,
- glass fibers: $SiO_2 – (Na_2O + K_2O) – (CaO + MgO) – (Al_2O_3 + Fe_2O_3)$,
- concretes and foam cement concretes as well as acoustical insulating composites: cements – (sand + + construction gypsum) – natural cellulose raw materials – polymeric binders.

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2. FORMATION OF TETRACOMPONENT STATE DIAGRAMS

Investigations have shown that the most optimal in all regards tetracomponent state diagram is a square.

Before the introduction of A, B, C, D amounts into the diagram, the group of four should be divided into two components pairs, e. g. A – B, C – D.

The sum of the components in the pair should be equal to 100 %.

As a result all the amounts of the components in the diagram are set on the square's sides and varies from 0 to 100 % as depicted in Fig. 1:

$$A\ (0 \div 100) \rightarrow C\ (0 \div 100) \rightarrow B\ (0 \div 100) \rightarrow D\ (0 \div 100)\ .$$

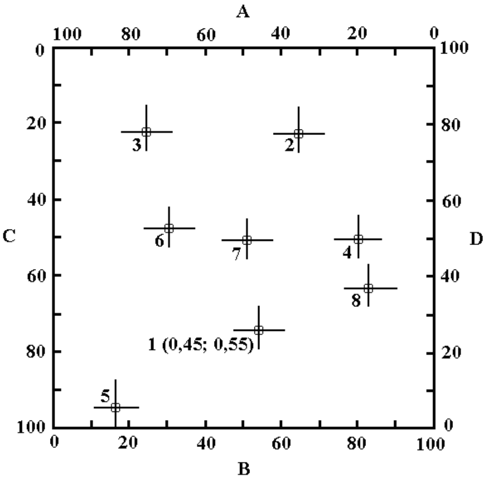

Fig. 1. Tetracomponent system A – B – C – D state diagram

2.1. Introduction of composite and material mixture composition into A tetracomponent diagram

This process is fulfilled as follows:

1) the amounts of A and B are determined so that its sum would be equal to 100 %;

2) the same is done with components C and D;

3) when the components A, B and its amounts are located on the sides, a line is drawn towards the square's center;

4) the same is preformed for C and D;

5) at the lines A – B and C – D interjunction, a dot is placed and marked, e. g. **K**.

Example

Given.

System **1** (%): A = 20, B = 25, C = 40, D = 15 (sum 100).

Task.

Introduce system **1** into a tetracomponent state diagram.

Solution.

1. We create two bicomponent systems:

1.1. A = 20, B = 25 (sum 45);
1.2. C= 40, D = 15 (sum 55); sum 100.

2. We reorganize these systems, so, that the components sum would be equal to 100 %.

2.1. We divide the amount of a A and B components by 0.45 (i. e. 45/100);

$A' = 20 : 0.45 = 44.5$; $B' = 25 : 0.45 = 55.5$ (sum 100).

2.2. We divide the amounts of C and D by 0.55 (i. e. 55/100):

$C' = 40 : 0.55 = 73$; $D' = 15 : 0.55 = 27$ (sum 100).

3. A′, B′, C′, D′ values are introduced into the diagram.

Result.

We mark the dot **1** in the diagram (Fig. 1).

Systems **2** – **8** are marked in a similar manner in Table 1.

Table 1. Data of 2 – 8 systems

	A	B	C	D	Sum
2	20	35	10	35	100
3	40	15	10	35	100
4	10	40	25	25	100
5	20	4	72	4	100
6	35	15	23	27	100
7	25	25	25	25	100
8	12	60	18	10	100

Note.

In order to be able to preform the opposite procedure – to ascertain the composition, it is purposive to demonstrate the dots number and composition factors: first of all – system A – B, after that system C – D. In this case the dot **1** should be marked as follows: **1** (**0.45**; **0.55**). One version exists, when the composition factors are not shown and are used in patent literature. In this case the composition factor values are patent "know-how".

2.2. Compound composition inclusion into the tetracomponent state diagram

Given.

Compound $6CaO \cdot Al_2O_3 \cdot 3SiO_2 \cdot 12H_2O$ (or $C_6AS_3H_{12}$) – belongs to the calcium alumosilicate family. Mole mass: CaO – 56, Al_2O_3 – 102, SiO_2 – 60, H_2O – 18. Compound mole mass M = 834.

Task.

Introduce the given compound composition into a tetracomponent $CaO – Al_2O_3 – SiO_2 – H_2O$ diagram (Fig. 2).

Solution.

1. Two bicomponent systems are created from the tetracomponent systems:

1) $CaO – SiO_2$, 2) $Al_2O_3 – H_2O$.

2. The amounts of CaO and SiO_2 in the material are found, %:

$$CaO = \frac{M_{6CaO}}{M} = \frac{336}{834} = 40.29\ \%\ ; \qquad (1)$$

$$SiO_2 = \frac{M_{3SiO_2}}{M} = \frac{180}{834} = 21.58\ \%\ ; \qquad (2)$$

$\Sigma_{CaO+SiO_2} = 40.29 + 21.58 = 61.87$ %, or 0.6187 parts. (3)

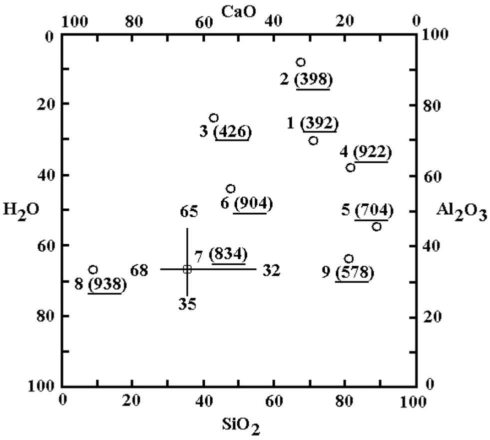

Fig. 2. Tetracomponent system $CaO - Al_2O_3 - SiO_2 - H_2O$ state diagram

3. The amount of Al_2O_3 and H_2O in the material, % are determined:

$$Al_2O_3 = \frac{M_{Al_2O_3}}{M} = \frac{102}{834} = 12.23\,\%\ ; \qquad (4)$$

$$H_2O = \frac{M_{12H_2O}}{M} = \frac{216}{834} = 25.90\,\%\ ; \qquad (5)$$

$\Sigma_{Al_2O_3+H_2O} = 12.23 + 25.90 = 38.13$ % or 0.3813 parts. (6)

Control:

61.87 (C) + 38.13 (F) = 100 %.

4. Correction of CaO and SiO_2 amounts, %:

CaO = 40.29 (A) : 0.6187 (C) = 65.12 % ; (7)

SiO_2 = 21.58 (B) : 0.6187 (C) = 65.12 % ; (8)

$\Sigma_{CaO+SiO_2}$ = 65.12 (K) + 34.88 (L) = 100 % .

5. Correction of Al_2O_3 and H_2O amounts, %:

Al_2O_3 = 12.23 (D) : 0.3813 (F) = 32.07 , (9)

H_2O = 25.90 (E) : 0.3813 (F) = 67.93 , (10)

$\Sigma_{Al_2O_3+H_2O}$ = 32.07 (M) + 67.93 (N) = 100 %

6. Fixation of the composition in the diagram (Fig. 2):

6.1. A vertical line is drawn at ~**65** % CaO (i. e. ~**35** % SiO_2), (in all 100 %).

6.2. A horizontal line is drawn at ~**32** % Al_2O_3 (i. e. ~**68** % H_2O), (in all 100 %).

7. At the intersection of the two segments, we show it as a dot, e. g. **41**. Besides the number, we can give the material's mole mass **834**, which will be necessary in identifying the dots composition.

Some other alumosilicate hydrate composition are given in Fig. 2 (M – mole mass):

1 – Scolecite CAS_3H_3

$CaO \cdot Al_2O_3 \cdot 3SiO_2 \cdot 3H_2O$ M = 392.

2 – Margarite CA_2S_2H

$CaO \cdot 2Al_2O_3 \cdot 2SiO_2 \cdot H_2O$ M = 398.

3 – Vesuvian $C_3AS_2H_2$

$3CaO \cdot Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$ M = 426.

4 – Leonhardite $C_2A_2S_8H_7$

$2CaO \cdot 2Al_2O_3 \cdot 8SiO_2 \cdot 7H_2O$ M = 922.

5 – Desmine CAS_7H_7

$CaO \cdot Al_2O_3 \cdot 7SiO_2 \cdot 7H_2O$ M = 704.

6 – Calciumhidroaliumosilicate $C_5A_2S_4H_{10}$

$5CaO \cdot 2Al_2O_3 \cdot 4SiO_2 \cdot 10H_2O$ M = 904.

7 – Calciumhidroaliumosilicate $C_6AS_3H_{12}$

$6CaO \cdot Al_2O_3 \cdot 3SiO_2 \cdot 12H_2O$ M = 834.

8 – Calciumhidroaliumosilicate $C_{10}ASH_{12}$

$10CaO \cdot Al_2O_3 \cdot SiO_2 \cdot 12H_2O$ M = 938.

9 – Calciumhidroaliumosilicate CAS_4H_{10}

$CaO \cdot Al_2O_3 \cdot 4SiO_2 \cdot 10H_2O$ M = 578.

3. THE ELUCIDATION OF THE SYSTEM'S COMPOSITION GIVEN IN THE TETRACOMPONENT DIAGRAM

Systems, in this case, can be made up of: 1) composites, 2) material mixtures, 3) chemical compounds. However, the identification of the system's composition differs among themselves.

3.1. Identification of composite and material mixture composition in tetracomponent state diagrams

Two versions are possible in this case:

1) when alongside the dot in the diagram with a serial number, other composition factor values are given. E. g. the dot in Fig. 1 is marked as **1** (**0.45**; **0.55**) (in the Fig. 1 **0.45** means the sum (A + B), divided by 100 %; by analogy the value **0.55** means the sum (C + D), divided by 100 %). (See part 2 *Solutions* 1.1 and 1.2);

2) when besides the dot's serial number, composition factor values are not given.

3.1.1. The elucidation of the system when composition factors are absent

Given.

System **1** (**0.45; 0.55**) system A – B – C – D state diagram (Fig. 1).

Task.

Find the systems A – B – C – D composition (%).

Solution.

1. The component amounts are evaluated, %:

1.1. According to the vertical line – A′ = 45; B′ = 55; (sum 100).

1.2. According to the horizontal line – C′ = 73; D′ = 27; (sum 100).

2. Then we multiply the obtained values by the dot's number sum factor value:

2.1. A = A′ · 0.45 = 20; B = B′ · 0.45 = 25.

2.2. C = C′ · 0.55 = 40; D = D′ · 0.55 = 15.

2.3. Sum 20 + 25 + 40 + 15 = 100 %.

3. Then we obtain the following system's composition: A = 20; B =25; C = 40; D = 15.

3.1.2. Elucidation of system's composition in absence of composition factor values

Given.

A system in which the corresponding composition is marked by a dot **M** (Fig. 3).

Task.

Find the system's **M** composition (%).

Solution.

1. Lines A_1B_1 and C_1D_1 are drawn through dot **M**.

2. The A_1, B_1, C_1, D_1 values are determined from the diagram:

$A_1 = 33.3$; $B_1 = 66.7$; $C_1 = 42.9$; $D_1 = 57.1$ (11)

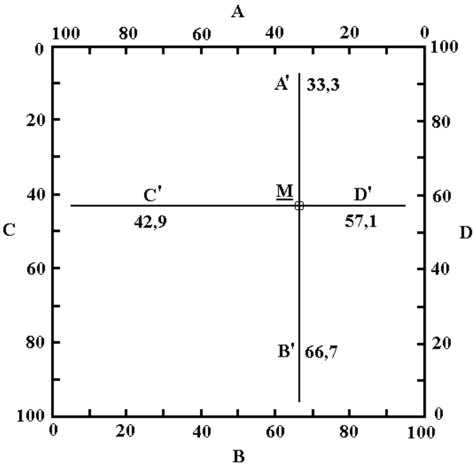

Fig. 3. Tetracomponent system A – B – C – D state diagram which contains two bicomponent system A – B and C – D

3. Two bicomponent system A – B and C – D are formed.

4. For these systems we have:

4.1. $A_1 = 100A / (A + B)$; (12)

$B_1 = 100B / (A + B)$; $(A_1 + B_1 = 100)$. (13)

4.2. $C_1 = 100C / (C + D)$; (14)

$D_1 = 100D / (C + D)$; (15) $(C_1 + D_1 = 100)$.

5. From (12), (13), (14), (15) we obtain:

$A = A_1(A + B) / 100$; (16)

$B = B_1(A + B) / 100$; (17)

$C = C_1(C + D) / 100$; (18)

$D = D_1(C + D) / 100$. (19)

6. By using A_1, B_1, C_1, D_1 values from (11), we find:

$A = 0.333 (A + B)$; $B = 0.667 (A + B)$. (20)

From here we obtain:

$B = 2A$; $C = 0.429 (C + D)$; $D = 0.571 (C + D)$. (21)

From here we obtain:

$D = 1.331C$

or: $A = 05B$; (22)

$C = 0.751D$. (23)

7. The (20) and (21) $C = f(A)$ values are inserted into the equation $A + B + C + D = 100$:

$A + 2A + C + 1.331C = 100$.

From here: $C = 42.882 – 1.286A$. (24)

8. The (20) and (23) $D = f(A)$ values are inserted into the equation $A + B + C + D = 100$:

$A + 2A + 0.751D + D = 100$;

$D = 57.11 – 1.713 A$. (25)

9. When we insert the (20), (24) and (25) values into equation $A + B + C + D = 100$ we obtain the identity $0 = 0$.

10. When we use the (20), (24) and (25) values, we draw up a corresponding one another A, B, C, D value from Table 2.

Table 2. A, B, C, D values

A	10	20	30	5	25
B = 2A	20	40	60	10	50
C = 42.882 – 1.286A	30	17	4	36	11
D = 57.11 – 1.713A	40	23	6	49	14
$A_1 + B_1 + C_1 + D_1 = 200$. A + B + C + D	100	100	100	100	100

As it can be seen from the Table 2; we obtain many solution combinations, all of which fit the dot **M** position.

11. Taking all of this into account, we need to adopt a rule to which we need to strictly abide with: when a dot **M** is included into a tetracomponent diagram, it is absolutely necessary to make a mark about the ratio $(A + B) / 100$ value at the dot.

As a result the elucidation of the composition at the dot's position in a tetracomponent system with a ratio $(A + B) / 100$ should be conducted first of all by performing the procedures in points 1 and 2, then by multiplying the obtained A_1 and B_1 values by $(A + B) / 100$, while the C_1 and D_1 – by $1 – (A + B)/100$.

Example

1. *Task.*

Find dot **M** (0.3)*, shown in Fig. 3, composition, if we know:

$A_1 = 33.3$; $B_1 = 66.7$; $C_1 = 42.9$; $D_1 = 57.1$;

$A_1 + B_1 + C_1 + D_1 = 200$.

Note*. Dot **M (0.3) can be ciphered by numbers, e. g. 6(0.4), 14(0.5), etc.

2. *Solution*:

2.1. A_1 and B_1 is multiplied by 0.3:

$A = 0.3\ A_1 = 0.3 \cdot 33.3 = 10$,

$B = 0.3\ B_1 = 0.3 \cdot 66.7 = 20$.

2.2. C_1 and D_1 values are multiplied by 0.7:

$C = 0.7\ C_1 = 0.7 \cdot 42.9 = 30$,

$D = 0.7\ D_1 = 0.7 \cdot 57.1 = 40$.

3. *Answer*:

$A = 10$; $B = 20$; $C = 30$; $D = 40$.

Sum $10 + 20 + 30 + 40 = 100$.

3.2. The elucidation of chemical compounds given in tetracomponent state diagrams

The chemical compound composition is ciphered by the dot given in the tetracomponent state diagram and is revealed by using a combination table.

Given.

A dot's number in the diagram (Fig. 2) and material's mole mass: No 41, **M** = **834**.

Solution.

1. The CaO, SiO_2, Al_2O_3 and H_2O amounts in the material are found graphically from the diagram according to the vertical and horizontal lines, %:

CaO 65; SiO_2 35 (sum 100);
Al_2O_3 32; H_2O 68 (sum 100).

2. Then we find the mole ratio from the diagram (molar masses: $CaO - 56$, $Al_2O_3 - 102$, $SiO_2 - 60$, $H_2O - 18$):

2.1. We find the $CaO - SiO_2$ mole ratio by the vertical direction:

$$\mathrm{CaO} : \mathrm{SiO_2} = \frac{65}{56} : \frac{35}{60} = 1.161 : 0.583 \cong 2:1,$$

i. e. $n\mathrm{C_2S}$.

2.2. We find $Al_2O_3 - H_2O$ molar ratio by the horizontal direction:

$$\mathrm{Al_2O_3} : \mathrm{H_2O} = \frac{32}{102} : \frac{68}{18} = 0.314 : 3.778 \cong 1:12,$$

i. e. m $\mathrm{AH_{12}}$.

3. We determine the dot's composition.

3.1. We draw up an equation system (834 – material molar mass):

$$n\,\mathrm{C_2S} + m\,\mathrm{AH_{12}} = 834,$$

$$n\,M_{\mathrm{C_2S}} + m\,\mathrm{M}_{\mathrm{AH_{12}}} = 834,$$

$$n\,(2M_{\mathrm{CaO}} + M_{\mathrm{SiO_2}}) + m\,(M_{\mathrm{Al_2O_3}} + 12\,M_{\mathrm{H_2O}}) = 834,$$

$$n\,(2\cdot 56 + 60) + m\,(102 + 12\cdot 18) = 834,$$

$$172\,n + 318\,m = 834.$$

3.2. n and m values are found from combination Table 3:

Table 3. *n* and *m* values

n \ m	1	2	3	4
1	490	808	1126	X
2	660	978	X	X
3	834	X	X	X
4	X	X	X	X

Answer $n = 3$ and $m = 1$. This represents the combination 3 $\mathrm{C_2S}$ and 1 $\mathrm{AH_{12}}$, i. e. $\mathrm{3C_2S \cdot AH_{12}}$ or $\mathrm{C_6AS_3H_{12}}$, finally $6\,\mathrm{CaO}\cdot\mathrm{Al_2O_3}\cdot 3\,\mathrm{SiO_2}\cdot 12\,\mathrm{H_2O}$.

4. CONCLUSIONS

Applying the method of tetracomponent system diagrams for analysis of materials compositions, we proposed new posibility for analysis of various combinations of chemical compounds (e. g. alumosilicate hydrate, etc.), rocks, construction materials, foam concretes, composites as well as raw material mixtures containing four main components. This diagram can have isotherms or other physico-chemical indices.

Method takes possibility: a) to include the chemical composition of various composites into diagrams, b) to elucidate the composition of systems in the diagram.

A possibility is foreseen how to include the chemical composition into the diagrams for use in patent material by ciphering it as "know-how" category data.

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