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Chemistry

SUBSOLIDUS STRUCTURE OF THE $MgO - Al_2O_3 - FeO - TiO_2$ SYSTEM

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Abstract. The subsolidus structure of the four-component system $MgO - Al_2O_3 - FeO - TiO_2$ was studied in six temperature ranges. Geometric-topological characteristics of the phases of the system under study have been determined, topological graphs of the relationship of elementary tetrahedrons have been constructed, their volumes, degrees of asymmetry for all temperature ranges have been found. The optimal regions of compositions for the production of spinel-containing materials have been predicted, which are within the limits of elementary tetrahedra: $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$, $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$, $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$ and $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$.

Keywords: subsolidus structure, tetrahedration, geometric-topological characteristics, magnesium-alumina spinel, hercynite, quandilite

1. Introduction

The four-component system $MgO - Al_2O_3 - FeO - TiO_2$ includes binary compounds such as spinels of various compositions, magnesium titanates and iron titanates, which are used to produce composite materials with valuable performance properties for various purposes. Particular attention should be paid to spinel binary compounds: magnesium-alumina spinel ($MgAl_2O_4$), hercynite ($FeAl_2O_4$), ulvospinel (Fe_2TiO_4) and quandilite (Mg_2TiO_4).

Magnesium-alumina spinel is one of the most important materials in the production of technical ceramics and refractories. Materials based on $MgAl_2O_4$ have a relatively high mechanical strength, high temperature resistance, good corrosion and radiation resistance.[1-3]

In recent years, the use of iron-alumina spinel (hercynite) for the lining of rotary kilns in the cement industry has been of increasing interest in the refractory industry.[4,5] These refractories having good coating properties are able to withstand thermal stress cracking, and are highly resistant to chemical corrosion.

Ceramics based on quandilite are widely used as dielectrics in resonators, filters and antennas for communications, radars operating at microwave frequencies.[6]

It is also possible to combine different types of spinels to create high performance materials. For example, the use of magnesia-alumina and iron-alumina spinels in the composition of the batch promotes a more intense formation of a uniform spider-web microporous structure due to different thermal expansion of these spinels.[7-9]

There is no information on the subsolidus structure of the four-component system $MgO - Al_2O_3 - FeO - TiO_2$ in the books, which causes certain difficulties in the synthesis of periclase-spinel refractories of a given phase composition with a set of predetermined operational properties. Thus, a detailed study of the subsolidus structure of the $MgO - Al_2O_3 - FeO - TiO_2$ system and its analysis is urgent.

Theoretical part

The subsolidus structure of the three-component systems: $Al_2O_3 - TiO_2 - FeO$, $MgO - TiO_2 - FeO$, $MgO - Al_2O_3 - FeO$, $MgO - Al_2O_3 - TiO_2$, which are part of the four component system under study, were examined by the authors in earlier publications.[10-12] According to studies of the thermodynamics of solid-phase exchange reactions, which limit the subsolidus structure of the four-component system $MgO - Al_2O_3 - FeO - TiO_2$,[13] the phase changes in this system occur in six temperature ranges: I – 800–1141 K, II – 1141–1413 K, III – 1413–1537 K, IV – 1537–1630 K, V – 1630–2076 K, VI – above 2076 K.

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This article is devoted to the test of the subsolidus structure of the four-component system $MgO – Al_2O_3 – FeO – TiO_2$. For clarity, the study of the system, its tetrahedration, which is described in detail in the publication,[13] will be presented in the form of figures (Fig. 1, 3, 5, 7, 9, 11). For each temperature interval the geometric-topological characteristics of the system and its phases were calculated: the volumes of elementary tetrahedra (V_i, ‰), the lengths of the conodes (Tables 1 – 6), the degree of their asymmetry (L_{max}/L_{min}) and the probability of the existence of phases (Tables 1-6) in accordance with the methods.[14] To describe the relationship between the elementary tetrahedrons of the system, topological graphs were made (Fig. 2, 4, 6, 8, 10) for each temperature interval. The verification of the correctness of the construction by the number of edges of the topological graph was carried out according to Euler formula.[14]

2. Results and Discussion

According to Fig. 1 concentration tetrahedron of the four-component system $MgO – Al_2O_3 – FeO – TiO_2$ in the temperature range of 800–1141 K has two "internal" conodes, which determines the presence of 10 elementary tetrahedra in it:

1) $Al_2O_3 – TiO_2 – FeTiO_3 – MgTi_2O_5$ (volume – 94 ‰, degree of asymmetry – 5.000),

2) $Al_2O_3 – FeTiO_3 – MgTi_2O_5 – MgTiO_3$ (volume – 63 ‰, degree of asymmetry – 6.895),

3) $Mg_2TiO_4 – FeAl_2O_4 – MgO – MgAl_2O_4$ (volume – 149 ‰, degree of asymmetry – 2.377),

4) $Mg_2TiO_4 – FeAl_2O_4 – MgO – FeO$ (volume – 293 ‰, degree of asymmetry – 2.000),

5) $Mg_2TiO_4 – FeAl_2O_4 – FeO – Fe_2TiO_4$ (volume – 105 ‰, degree of asymmetry – 2.426),

6) $FeAl_2O_4 – MgTiO_3 – MgAl_2O_4 – Mg_2TiO_4$ (volume – 50 ‰, degree of asymmetry – 4.383),

7) $FeAl_2O_4 – MgTiO_3 – Mg_2TiO_4 – Fe_2TiO_4$ (volume – 63 ‰, degree of asymmetry – 4.383),

8) $FeAl_2O_4 – MgTiO_3 – Fe_2TiO_4 – FeTiO_3$ (volume – 33 ‰, degree of asymmetry – 4.331),

9) $FeAl_2O_4 – MgTiO_3 – MgAl_2O_4 – Al_2O_3$ (volume – 78 ‰, degree of asymmetry – 3.139),

10) $FeAl_2O_4 – MgTiO_3 – Al_2O_3 – FeTiO_3$ (volume – 72 ‰, degree of asymmetry – 2.092).

In this temperature range, the tetrahedron $Mg_2TiO_4 – FeAl_2O_4 – MgO – FeO$ has the largest volume and the least degree of asymmetry. The tetrahedra $Mg_2TiO_4 – FeAl_2O_4 – MgO – MgAl_2O_4$ and $Mg_2TiO_4 – FeAl_2O_4 – FeO – Fe_2TiO_4$ also have relatively large volumes and low degrees of asymmetry. These three tetrahedra include phases that are important for periclase-spinel refractories: hercynite (has the highest probability of existence (Table 1) and coexists with the greatest number of phases), magnesium-alumina spinel (the probability of existence is average compared to other compounds), and quandilite (the probability of existence is relatively high).

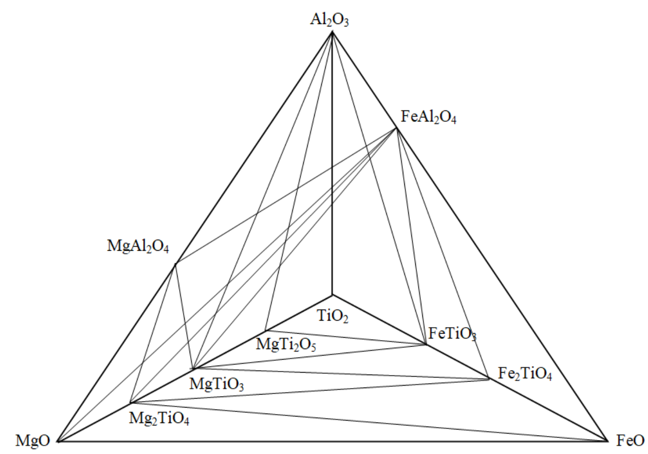

Fig. 1. Subsolidus structure of the $MgO – Al_2O_3 – FeO – TiO_2$ system in the temperature range of 800–1141 K

In Fig. 2 the topological graph of the interconnection of elementary tetrahedra of the system is shown; it is flat and without plug-in tetrahedra. According to Euler's formula, the number of edges of a topological graph (R) is 11, which proves the correctness of the graph construction. The "hanging" vertex of the graph corresponds to the first of the indicated elementary tetrahedra, which means that three of its four faces emerge on the edge of the concentration tetrahedron of the $MgO – Al_2O_3 – FeO – TiO_2$ system. Three vertices of the graph have a degree of connectivity of 3, that is for tetrahedra 6, 7, and 10, only one face goes to some face of the concentration tetrahedron of the system.

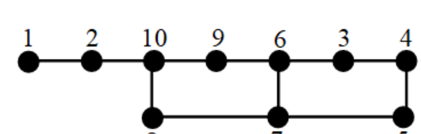

Fig. 2. Topological graph of the relationship of elementary tetrahedra of the $MgO – Al_2O_3 – FeO – TiO_2$ system in the temperature range of 800 – 1141 K

Table 1. Geometric-topological characteristics of the phases of the MgO – Al_2O_3 – FeO – TiO_2 system in the temperature range of 800–1141 K

Phase	Coexisting phases and lengths of conodes, L, ‰	Number of elementary tetrahedra, where the phase exists	Total volume of existence, ‰	Probability of existence, ω, fractions of units
MgO	FeO (1000), $FeAl_2O_4$ (870), $MgAl_2O_4$ (719), Mg_2TiO_4 (500)	2	442	0.1105
Al_2O_3	TiO_2 (1000), $MgTi_2O_5$ (917), $FeTiO_3$ (866), $MgTiO_3$ (882), $MgAl_2O_4$ (281), $FeAl_2O_4$ (414),	4	307	0.0767
FeO	MgO (1000), Mg_2TiO_4 (866), $FeAl_2O_4$ (586), Fe_2TiO_4 (357)	2	395	0,0995
TiO_2	Al_2O_3 (1000), $FeTiO_3$ (474), $MgTi_2O_5$ (200)	1	94 (min)	0.0235 (min)
$MgAl_2O_4$	MgO (719), $MgTiO_3$ (694), Mg_2TiO_4 (638), $FeAl_2O_4$ (366), Al_2O_3 (281)	3	277	0.0693
$FeAl_2O_4$	MgO (870), $MgTiO_3$ (732), Mg_2TiO_4 (712), FeO (586), $FeTiO_3$ (558), Fe_2TiO_4 (512), Al_2O_3 (414), $MgAl_2O_4$ (366)	8	843 (max)	0.2108 (max)
Fe_2TiO_4	Mg_2TiO_4 (585), $MgTiO_3$ (555), $FeAl_2O_4$ (512), FeO (357), $FeTiO_3$ (169)	3	201	0.0503
$FeTiO_3$	Al_2O_3 (866), $FeAl_2O_4$ (558), TiO_2 (474), $MgTiO_3$ (422), $MgTi_2O_5$ (412), Fe_2TiO_4 (169)	4	262	0.0655
$MgTi_2O_5$	Al_2O_3 (917), $FeTiO_3$ (412), TiO_2 (200), $MgTiO_3$ (133)	2	157	0.0392
$MgTiO_3$	Al_2O_3 (882), $FeAl_2O_4$ (732), $MgAl_2O_4$ (694), Fe_2TiO_4 (555), $FeTiO_3$ (422), Mg_2TiO_4 (167), $MgTi_2O_5$ (133)	6	359	0.0897
Mg_2TiO_4	FeO (866), $FeAl_2O_4$ (712), $MgAl_2O_4$ (638), Fe_2TiO_4 (585), MgO (500), $MgTiO_3$ (167)	5	660	0.1650
	Σ		4000	1.0000

When dividing the concentration tetrahedron into elementary tetrahedra in the temperature range of 1141–1413 K (Fig. 3), two “internal” conodes were found, which determine the presence of 10 elementary tetrahedra in the subsolidus structure of the system under study:

1) Al_2O_3 – TiO_2 – $FeTiO_3$ – $MgTi_2O_5$ (volume – 94 ‰, degree of asymmetry – 5.000),

2) Al_2O_3 – $FeTiO_3$ – $MgTi_2O_5$ – $MgTiO_3$ (volume – 63 ‰, degree of asymmetry – 6.895),

3) MgO – $MgAl_2O_4$ – Mg_2TiO_4 – FeO (volume – 360 ‰, degree of asymmetry – 2.000).

4) $FeAl_2O_4$ – Mg_2TiO_4 – $MgAl_2O_4$ – FeO (volume – 82 ‰, degree of asymmetry – 2.440),

5) $FeAl_2O_4$ – Mg_2TiO_4 – FeO – Fe_2TiO_4 (volume – 105 ‰, degree of asymmetry – 2.426),

6) $FeAl_2O_4$ – Mg_2TiO_4 – Fe_2TiO_4 – $FeTiO_3$ (volume – 49 ‰, degree of asymmetry – 4.213),

7) Mg_2TiO_4 – $MgTiO_3$ – $FeTiO_3$ – $FeAl_2O_4$ (volume – 46 ‰, degree of asymmetry – 4.383),

8) $FeAl_2O_4$ – $MgTiO_3$ – $MgAl_2O_4$ – Mg_2TiO_4 (volume – 50 ‰, degree of asymmetry – 4.383),

9) $FeAl_2O_4$ – $MgTiO_3$ – $MgAl_2O_4$ – Al_2O_3 (volume – 78 ‰, degree of asymmetry – 3.139),

10) $FeAl_2O_4$ – $MgTiO_3$ – Al_2O_3 – $FeTiO_3$ (volume – 72 ‰, degree of asymmetry – 2.092).

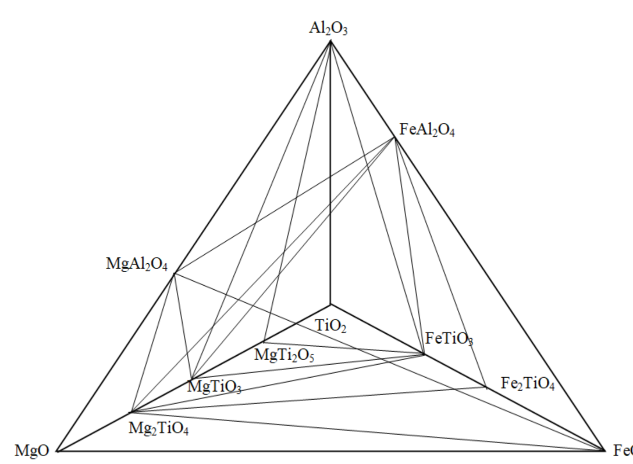

Fig. 3. Subsolidus structure of the MgO – Al_2O_3 – FeO – TiO_2 system in the temperature range of 1141 – 1413 K

In the temperature range of 1141–1413 K, the tetrahedron MgO – $MgAl_2O_4$ – Mg_2TiO_4 – FeO has the largest volume and the least degree of asymmetry. Tetrahedrons $FeAl_2O_4$ – Mg_2TiO_4 – FeO – Fe_2TiO_4, Al_2O_3 – TiO_2 – $FeTiO_3$ – $MgTi_2O_5$, $FeAl_2O_4$ – Mg_2TiO_4 – $MgAl_2O_4$ – FeO, $FeAl_2O_4$ – $MgTiO_3$ – $MgAl_2O_4$ – Al_2O_3 and $FeAl_2O_4$ – $MgTiO_3$ – Al_2O_3 – $FeTiO_3$ have comparatively high volume and average degrees of asymmetry, except the tetrahedron $Al_2O_3 – TiO_2 – FeTiO_3 – MgTi_2O_5$, which has a high degree of asymmetry.

These tetrahedrons contain various types of spinels. Quandilite has the highest probability of existence (Table 2) and coexists with the largest number of phases, followed by a hercynite and magnesium-alumina spinel in the descending order.

The topological graph of the interconnection of the elementary tetrahedra of the system for a given temperature interval (Fig. 4) is flat and without plug-in tetrahedrons, but it is more complexly organized in the central part: two “hanging” points and four vertices with a degree of connectivity 3. According to Euler's formula, the number of edges of the topological graph (R) is equal to 11, which proves the correctness of the graph construction.

Table 2. Geometric-topological characteristics of the phases of the system $MgO – Al_2O_3 – FeO – TiO_2$ in the temperature range 1141–1413 K

Phase	Coexisting phases and lengths of conodes, L, ‰	Number of elementary tetrahedra, where the phase exists	Total volume of existence, ‰	Probability of existence, ω, fractions of units
MgO	FeO (1000), $MgAl_2O_4$ (719), Mg_2TiO_4 (500)	1	361	0.0903
Al_2O_3	TiO_2 (1000), $MgTi_2O_5$ (917), $FeTiO_3$ (866), $MgTiO_3$ (882), $FeAl_2O_4$ (414), $MgAl_2O_4$ (281)	4	307	0.0767
FeO	MgO (1000), $MgAl_2O_4$ (893), Mg_2TiO_4 (866), $FeAl_2O_4$ (586), Fe_2TiO_4 (357)	3	548	0.1370
TiO_2	Al_2O_3 (1000), $FeTiO_3$ (474), $MgTi_2O_5$ (200)	1	94 (min)	0.0235 (min)
$MgAl_2O_4$	FeO (893), MgO (719), $MgTiO_3$ (694), Mg_2TiO_4 (638), $FeAl_2O_4$ (366), Al_2O_3 (281)	4	571	0.1428
$FeAl_2O_4$	$MgTiO_3$ (732), Mg_2TiO_4 (712), Al_2O_3 (414), FeO (586), $FeTiO_3$ (558), Fe_2TiO_4 (512), $MgAl_2O_4$ (366)	7	482	0.1205
Fe_2TiO_4	Mg_2TiO_4 (585), $FeAl_2O_4$ (512), FeO (357), $FeTiO_3$ (169)	2	154	0.0385
$FeTiO_3$	Al_2O_3 (866), $FeAl_2O_4$ (558), Mg_2TiO_4 (488), TiO_2 (474), $MgTiO_3$ (422), $MgTi_2O_5$ (412), Fe_2TiO_4 (169)	5	324	0.0810
$MgTi_2O_5$	Al_2O_3 (917), $FeTiO_3$ (412), TiO_2 (200), $MgTiO_3$ (133)	2	157	0.0392
$MgTiO_3$	Al_2O_3 (882), $FeAl_2O_4$ (732), $MgAl_2O_4$ (694), $FeTiO_3$ (422), Mg_2TiO_4 (167), $MgTi_2O_5$ (133)	5	309	0.0773
Mg_2TiO_4	FeO (866), $FeAl_2O_4$ (712), $MgAl_2O_4$ (638), Fe_2TiO_4 (585), MgO (500), $FeTiO_3$ (488), $MgTiO_3$ (167)	6	693 (max)	0.1732 (max)
$\sum$			4000	1.0000

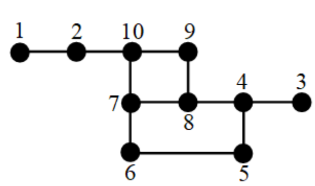

Fig. 4. Topological graph of the relationship of elementary tetrahedra of the $MgO – Al_2O_3 – FeO – TiO_2$ system in the temperature range of 1141–1413 K

In the temperature range of 1413–1537 K, as a result of the division of the concentration tetrahedron into elementary tetrahedra (Fig. 5), three “internal” conodes were found, which determine the presence of 12 elementary tetrahedra in the subsolidus structure of the system under study:

1) $MgO – FeO – Mg_2TiO_4 – MgAl_2O_4$ (volume – 360 ‰, degree of asymmetry – 2.000),

2) $FeAl_2O_4 – Mg_2TiO_4 – MgAl_2O_4 – FeO$ (volume – 82 ‰, degree of asymmetry – 2.440),

3) $FeAl_2O_4 – Mg_2TiO_4 – FeO – Fe_2TiO_4$ (volume – 105 ‰, degree of asymmetry – 2.426),

4) $FeAl_2O_4 – Mg_2TiO_4 – Fe_2TiO_4 – FeTiO_3$ (volume – 49 ‰, degree of asymmetry – 4.213),

5) $FeAl_2O_4 – MgTiO_3 – Mg_2TiO_4 – MgAl_2O_4$ (volume – 50 ‰, degree of asymmetry – 4.383),

6) $FeAl_2O_4 – MgTiO_3 – Mg_2TiO_4 – FeTiO_3$ (volume – 46 ‰, degree of asymmetry – 4.383),

7) $FeAl_2O_4 – MgTiO_3 – MgAl_2O_4 – Al_2O_3$ (volume – 78 ‰, degree of asymmetry – 3.139),

8) $FeAl_2O_4 – MgTi_2O_5 – MgTiO_3 – Al_2O_3$ (volume – 55 ‰, degree of asymmetry – 6.895),

9) $FeAl_2O_4 - MgTi_2O_5 - MgTiO_3 - FeTiO_3$(volume – 37 ‰, degree of asymmetry – 5.812),

10) $FeAl_2O_4 - MgTi_2O_5 - FeTiO_3 - FeTi_2O_5$ (volume – 19 ‰, degree of asymmetry – 4.713),

11) $FeAl_2O_4 - MgTi_2O_5 - FeTi_2O_5 - TiO_2$ (volume – 36 ‰, degree of asymmetry – 4.350),

12) $FeAl_2O_4 - MgTi_2O_5 - TiO_2 - Al_2O_3$ (volume – 83 ‰, degree of asymmetry – 5.000).

In this temperature range, the elementary tetrahedron $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$ has the largest volume, as in the previous temperature range, and the least degree of asymmetry. For elementary tetrahedra $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$, $FeAl_2O_4 - MgTi_2O_5 - TiO_2 - Al_2O_3$, $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$ и $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$ the average volume is relatively high with average degrees of asymmetry, except for $FeAl_2O_4 - MgTi_2O_5 - TiO_2 - Al_2O_3$ which has a high degree of asymmetry. These tetrahedrons also include various types of spinels. Quandilite has the highest probability of existence (Table 3) and coexists with the largest number of phases, then hercynite in a descending order, and then the magnesium-alumina spinel.

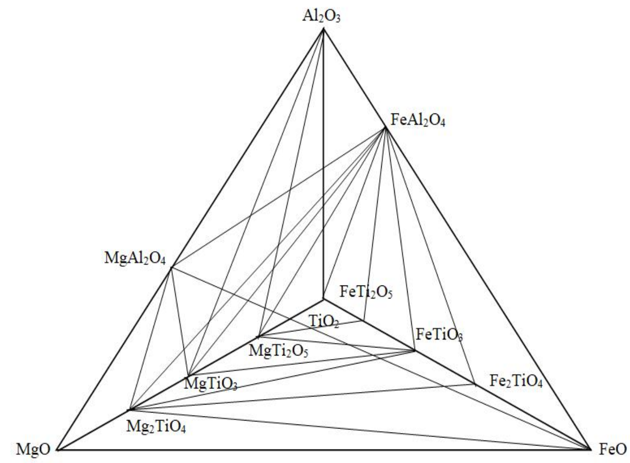

Fig. 5. Subsolidus structure of the $MgO - Al_2O_3 - FeO - TiO_2$ system in the temperature range of 1413–1537 K

Table 3. Geometric-topological characteristics of the phases of the system$MgO - Al_2O_3 - FeO - TiO_2$ in the temperature range of 1413–1537 K

Phase	Coexisting phases and lengths of conodes, L, ‰	Number of elementary tetrahedra, where the phase exists	Total volume of existence, ‰	Probability of existence, ω, fractions of units
MgO	FeO (1000), $MgAl_2O_4$ (719), Mg_2TiO_4 (500)	1	360	0.0900
Al_2O_3	TiO_2 (1000), $MgTi_2O_5$ (917), $MgTiO_3$ (882), $FeAl_2O_4$ (414), $MgAl_2O_4$ (281)	3	216	0.0540
FeO	MgO (1000), $MgAl_2O_4$ (893), Mg_2TiO_4 (866), $FeAl_2O_4$ (586), Fe_2TiO_4 (357)	3	547	0.1367
TiO_2	Al_2O_3 (1000), $FeAl_2O_4$ (870), $FeTi_2O_5$ (310), $MgTi_2O_5$ (200)	2	119	0.0297
$MgAl_2O_4$	FeO (893), MgO (719), $MgTiO_3$ (694), Mg_2TiO_4 (638), $FeAl_2O_4$ (366), Al_2O_3 (281)	4	570	0.1425
$FeAl_2O_4$	TiO_2 (870), $MgTi_2O_5$ (773), $MgTiO_3$ (732), Mg_2TiO_4 (712), $FeTi_2O_5$ (644), FeO (586), $FeTiO_3$ (558), Fe_2TiO_4 (512), Al_2O_3 (414), $MgAl_2O_4$ (366)	11	640	0.1600
Fe_2TiO_4	Mg_2TiO_4 (585), $FeAl_2O_4$ (512), FeO (357), $FeTiO_3$ (169)	2	154	0.0385
$FeTiO_3$	$FeAl_2O_4$ (558), Mg_2TiO_4 (488), $MgTiO_3$ (422), $MgTi_2O_5$ (412), Fe_2TiO_4 (169), $FeTi_2O_5$ (164)	4	151	0.0378
$FeTi_2O_5$	$FeAl_2O_4$ (644), TiO_2 (310), $MgTi_2O_5$ (272), $FeTiO_3$ (164)	2	55 (min)	0.0138 (min)
$MgTi_2O_5$	Al_2O_3 (917), $FeAl_2O_4$ (773), $FeTiO_3$ (412), $FeTi_2O_5$ (272),TiO_2 (200), $MgTiO_3$ (133)	5	230	0.0575
$MgTiO_3$	Al_2O_3 (882), $FeAl_2O_4$ (732), $MgAl_2O_4$ (694), $FeTiO_3$ (422), Mg_2TiO_4 (167), $MgTi_2O_5$ (133)	5	266	0.0665
Mg_2TiO_4	FeO (866), $FeAl_2O_4$ (712), $MgAl_2O_4$ (638), Fe_2TiO_4 (585), MgO (500), $FeTiO_3$ (488), $MgTiO_3$ (167)	6	692 (max)	0.1730 (max)
$\sum$			4000	1.0000

The topological graph of the interconnection of elementary tetrahedra of the system for a given temperature interval (Fig. 6) is even more complicated: one "hanging" point and four vertices with a degree of connectivity 3. According to Euler's formula, the number of edges of the topological graph (R) is 14, which proves the correctness of the graph construction.

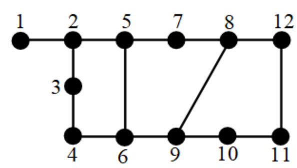

Fig. 6. Topological graph of the relationship of elementary tetrahedra of the $MgO – Al_2O_3 – FeO – TiO_2$ system in the temperature range of 1413–1537 K

In the temperature range of 1537 – 1630 K (Fig. 7), there are three "internal" conodes, which determine the presence of 13 elementary tetrahedra in the subsolidus structure of the $MgO – Al_2O_3 – FeO – TiO_2$ system:

1) $MgO – FeO – Mg_2TiO_4 – MgAl_2O_4$ (volume – 360 ‰, degree of asymmetry – 2.000),

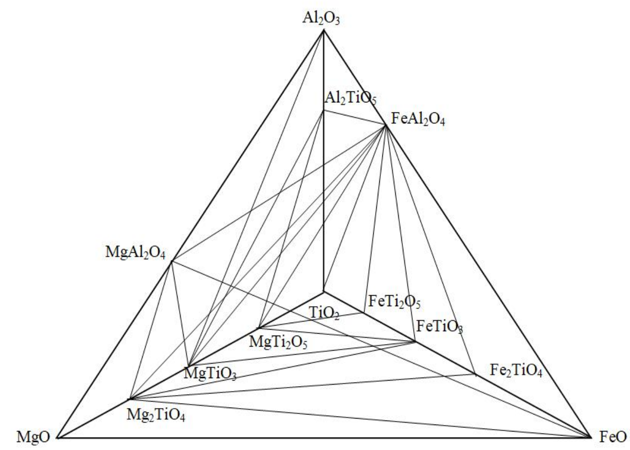

Fig. 7. Subsolidus structure of the $MgO – Al_2O_3 – FeO – TiO_2$ system in the temperature range of 1537–1630 K

Table 4. Geometric-topological characteristics of the phases of the system $MgO – Al_2O_3 – FeO – TiO_2$ in the temperature range of 1537–1630 K

Phase	Coexisting phases and lengths of conodes, L, ‰	Number of elementary tetrahedra, where the phase exists	Total volume of existence, ‰	Probability of existence, ω, fractions of units
MgO	FeO (1000), $MgAl_2O_4$ (719), Mg_2TiO_4 (500)	1	360	0.0900
Al_2O_3	$MgTiO_3$ (882), Al_2TiO_5 (440), $FeAl_2O_4$ (414), $MgAl_2O_4$ (281)	2	139	0.0347
FeO	MgO (1000), $FeAl_2O_4$ (586), Fe_2TiO_4 (357), Mg_2TiO_4 (866), $MgAl_2O_4$ (893)	3	547	0.1367
TiO_2	$FeAl_2O_4$ (870), Al_2TiO_5 (560), $FeTi_2O_5$ (310), $MgTi_2O_5$ (200)	2	82	0.0205
$MgAl_2O_4$	FeO (893), MgO (719), $MgTiO_3$ (694), Mg_2TiO_4 (638), $FeAl_2O_4$ (366), Al_2O_3 (281)	4	570	0.1425
Al_2TiO_5	TiO_2 (560), $MgTi_2O_5$ (492), $MgTiO_3$ (488), Al_2O_3 (440), $FeAl_2O_4$ (428)	3	138	0.0345
$FeAl_2O_4$	TiO_2 (870), $MgTi_2O_5$ (773), $MgTiO_3$ (732), Mg_2TiO_4 (712), $FeTi_2O_5$ (644), FeO (586), $FeTiO_3$ (558), Fe_2TiO_4 (512), Al_2TiO_5 (428), Al_2O_3 (414), $MgAl_2O_4$ (366)	12	640	0.1600
Fe_2TiO_4	Mg_2TiO_4 (585), $FeAl_2O_4$ (512), FeO (357), $FeTiO_3$ (169)	2	154	0.0385
$FeTiO_3$	$FeAl_2O_4$ (558), Mg_2TiO_4 (488), $MgTiO_3$ (422), $MgTi_2O_5$ (412), Fe_2TiO_4 (169), $FeTi_2O_5$ (164)	4	151	0.0378
$FeTi_2O_5$	$FeAl_2O_4$ (644), TiO_2 (310), $MgTi_2O_5$ (272), $FeTiO_3$ (164)	2	55 (min)	0.0138 (min)
$MgTi_2O_5$	$FeAl_2O_4$ (773), Al_2TiO_5 (492), $FeTiO_3$ (412), $FeTi_2O_5$ (272), TiO_2 (200), $MgTiO_3$ (133)	5	169	0.0423
$MgTiO_3$	Al_2O_3 (882), $FeAl_2O_4$ (732), $MgAl_2O_4$ (694), Al_2TiO_5 (488), $FeTiO_3$ (422), Mg_2TiO_4 (167), $MgTi_2O_5$ (133)	6	303	0.0757
Mg_2TiO_4	FeO (866), $FeAl_2O_4$ (712), $MgAl_2O_4$ (638), Fe_2TiO_4 (585), MgO (500), $FeTiO_3$ (488), $MgTiO_3$ (167)	6	692 (max)	0.1730 (max)
∑			4000	1.0000

2) $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$ (volume – 105 ‰, degree of asymmetry – 2.426),

3) $FeAl_2O_4 - Mg_2TiO_4 - Fe_2TiO_4 - FeTiO_3$ (volume – 49 ‰, degree of asymmetry – 4.213),

4) $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$ (volume – 82 ‰, degree of asymmetry – 2.440),

5) $FeAl_2O_4 - MgTiO_3 - Al_2TiO_5 - Al_2O_3$ (volume – 61 ‰, degree of asymmetry – 2.000),

6) $FeAl_2O_4 - MgTiO_3 - Mg_2TiO_4 - MgAl_2O_4$ (volume – 50 ‰, degree of asymmetry – 4.383),

7) $FeAl_2O_4 - MgTiO_3 - Mg_2TiO_4 - FeTiO_3$ (volume – 46 ‰, degree of asymmetry – 4.383),

8) $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$ (volume – 78 ‰, degree of asymmetry – 3.139),

9) $FeAl_2O_4 - MgTi_2O_5 - MgTiO_3 - Al_2TiO_5$ (volume – 31 ‰, degree of asymmetry – 5.812),

10) $FeAl_2O_4 - MgTi_2O_5 - Al_2TiO_5 - TiO_2$ (volume – 46 ‰, degree of asymmetry – 4.350),

11) $FeAl_2O_4 - MgTi_2O_5 - FeTiO_3 - FeTi_2O_5$ (volume – 19 ‰, degree of asymmetry – 4.713),

12) $FeAl_2O_4 - MgTi_2O_5 - TiO_2 - FeTi_2O_5$ (volume – 36 ‰, degree of asymmetry – 4.350),

13) $FeAl_2O_4 - MgTi_2O_5 - MgTiO_3 - FeTiO_3$ (volume – 37 ‰, degree of asymmetry – 5.812).

In this interval, the tetrahedron $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$ has the largest volume, as well as in the previous temperature intervals, and the least degree of asymmetry. The tetrahedra $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$, $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$, $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$ have relatively high volumes and average degrees of symmetry. The composition of these tetrahedra includes different types of spinels – qu-andilite has the highest probability of existence (Table 4) and coexists with the largest number of phases, then hercynite appears in the descending order, and then the magnesium-alumina spinel does.

The topological graph of the interconnection of elementary tetrahedra of the system for a given temperature interval (Fig. 8) is flat and without plug-in tetrahedra, but its structure is even more complex: one "hanging" point and five vertices with a degree of connectivity 3. According to Euler's formula, the number of edges of a topological graph (R) is equal to 15, which proves the correctness of the graph construction.

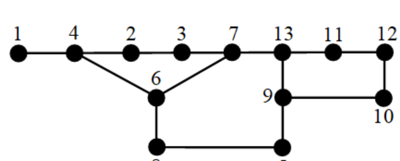

Fig. 8. Topological graph of the relationship of elementary tetrahedra of the $MgO - Al_2O_3 - FeO - TiO_2$ system in the temperature range of 1537 – 1630 K

In Fig. 9, the presence of two "internal" conodes in the subsolidus structure of the system under study was found in the temperature range of 1630–2076 K and the concentration tetrahedron is divided into 12 elementary tetrahedra:

1) $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$ (volume – 360 ‰, degree of asymmetry – 2.000),

2) $Al_2TiO_5 - TiO_2 - MgTi_2O_5 - FeTi_2O_5$ (volume – 35 ‰, degree of asymmetry – 2.800),

3) $Al_2TiO_5 - MgTi_2O_5 - FeTi_2O_5 - FeTiO_3$ (volume – 18 ‰, degree of asymmetry – 3.183),

4) $Al_2TiO_5 - MgTi_2O_5 - FeTiO_3 - MgTiO_3$ (volume – 35 ‰, degree of asymmetry – 3.925),

5) $Al_2TiO_5 - MgTiO_3 - FeTiO_3 - Al_2O_3$ (volume – 69 ‰, degree of asymmetry – 2.000),

6) $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$ (volume – 78 ‰, degree of asymmetry – 3.139),

7) $FeAl_2O_4 - MgTiO_3 - Al_2O_3 - FeTiO_3$ (volume – 73 ‰, degree of asymmetry – 2.130),

8) $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - MgTiO_3$ (volume – 50 ‰, degree of asymmetry – 4.383),

9) $FeAl_2O_4 - Mg_2TiO_4 - MgTiO_3 - FeTiO_3$ (volume – 46 ‰, degree of asymmetry – 4.383),

10) $FeAl_2O_4 - Mg_2TiO_4 - FeTiO_3 - Fe_2TiO_4$ (volume – 49 ‰, degree of asymmetry – 4.213),

11) $FeAl_2O_4 - Mg_2TiO_4 - Fe_2TiO_4 - FeO$ (volume – 105 ‰, degree of asymmetry – 2.426),

12) $FeAl_2O_4 - Mg_2TiO_4 - FeO - MgAl_2O_4$ (volume – 82 ‰, degree of asymmetry – 2.440).

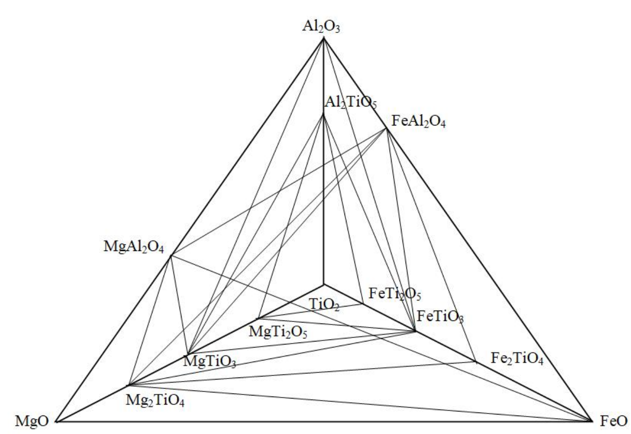

Fig. 9. Subsolidus structure of the $MgO - Al_2O_3 - FeO - TiO_2$ system in the temperature range of 1630 – 2076 K

In the temperature range under study, the tetrahedron $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$, has the largest volume, as well as in the previous temperature ranges, and the least degree of asymmetry. The tetrahedra $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$, $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$, $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$ and $FeAl_2O_4 - MgTiO_3 - Al_2O_3 - FeTiO_3$ have comparatively high volumes and average degrees of asymmetry (Table 5).

Table 5. Geometric-topological characteristics of the phases of the system $MgO – Al_2O_3 – FeO – TiO_2$ in the temperature range of 1630–2076 K

Phase	Coexisting phases and lengths of conodes, L, ‰	Number of elementary tetrahedra, where the phase exists	Total volume of existence, ‰	Probability of existence, ω, fractions of units
MgO	FeO (1000), $MgAl_2O_4$ (719), Mg_2TiO_4 (500)	1	360	0.0900
Al_2O_3	$MgTiO_3$ (882), $FeTiO_3$ (866), Al_2TiO_5 (440), $FeAl_2O_4$ (414), $MgAl_2O_4$ (281)	3	220	0.0550
FeO	MgO (1000), $MgAl_2O_4$ (893), Mg_2TiO_4 (866), $FeAl_2O_4$ (586), Fe_2TiO_4 (357)	3	547	0.1367
TiO_2	Al_2TiO_5 (560), $FeTi_2O_5$ (310), $MgTi_2O_5$ (200)	1	35	0.0087
$MgAl_2O_4$	FeO (893), MgO (719), $MgTiO_3$ (694), Mg_2TiO_4 (638), $FeAl_2O_4$ (366), Al_2O_3 (281)	4	570	0.1425
Al_2TiO_5	TiO_2 (560), $FeTiO_3$ (522), $MgTi_2O_5$ (492), $MgTiO_3$ (488), $FeTi_2O_5$ (486), Al_2O_3 (440)	4	157	0.0393
$FeAl_2O_4$	$MgTi_2O_5$ (773), $MgTiO_3$ (732), Mg_2TiO_4 (712), FeO (586), $FeTiO_3$ (558), Fe_2TiO_4 (512), Al_2O_3 (414), $MgAl_2O_4$ (366)	7	483	0.1208
Fe_2TiO_4	Mg_2TiO_4 (585), $FeAl_2O_4$ (512), FeO (357), $FeTiO_3$ (169)	2	154	0.0385
$FeTiO_3$	Al_2O_3 (866), $FeAl_2O_4$ (558), Al_2TiO_5 (522), Mg_2TiO_4 (488), $MgTiO_3$ (422), $MgTi_2O_5$ (412), Fe_2TiO_4 (169), $FeTi_2O_5$ (164)	6	290	0.0725
$FeTi_2O_5$	Al_2TiO_5 (486), TiO_2 (310), $MgTi_2O_5$ (272), $FeTiO_3$ (164)	2	53	0.0133
$MgTi_2O_5$	$FeAl_2O_4$ (773), Al_2TiO_5 (492), $FeTiO_3$ (412), $FeTi_2O_5$ (272), TiO_2 (200), $MgTiO_3$ (133)	3	88	0.0220
$MgTiO_3$	Al_2O_3 (882), $FeAl_2O_4$ (732), $MgAl_2O_4$ (694), Al_2TiO_5 (488), $FeTiO_3$ (422), Mg_2TiO_4 (167), $MgTi_2O_5$ (133)	6	351	0.0877
Mg_2TiO_4	FeO (866), $FeAl_2O_4$ (712), $MgAl_2O_4$ (638), Fe_2TiO_4 (585), MgO (500), $FeTiO_3$ (488), $MgTiO_3$ (167)	6	692	0.1730
∑			4000	1.0000

The topological graph of the interconnection of elementary tetrahedra of the system for a given temperature interval (Fig. 10) is flat and without plug-in tetrahedrons, there are two "hanging" points and four vertices with a degree of connectivity 3. According to Euler's formula, the number of edges of the topological graph (R) is 13, which proves the correctness of the construction of the graph.

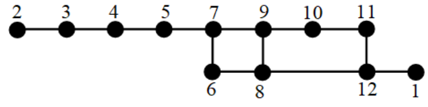

Fig. 10. Topological graph of the relationship of elementary tetrahedra of the $MgO – Al_2O_3 – FeO – TiO_2$ system in the temperature range of 1630–2076 K

Above the temperature of 2076 K (Fig. 11), elementary tetrahedrons are automatically closed in the concentration tetrahedron of the system under study:

1) $Al_2O_3 – Al_4TiO_8 – FeAl_2O_4 – MgAl_2O_4$ (volume – 32 ‰, degree of asymmetry – 1.437),

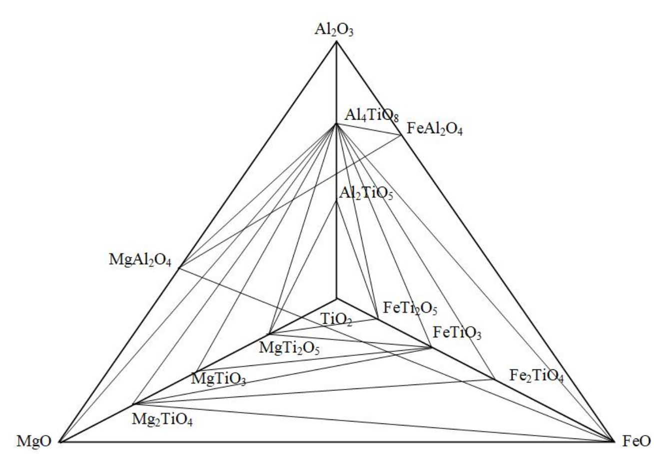

Fig. 11. Subsolidus structure of the $MgO – Al_2O_3 – FeO – TiO_2$ system above a temperature of 2076 K

2) Al_4TiO_8 – $FeAl_2O_4$ – $MgAl_2O_4$ – FeO (volume – 46 ‰, degree of asymmetry – 3.167),

3) Al_4TiO_8 – $MgAl_2O_4$ – MgO – FeO (volume – 203 ‰, degree of asymmetry – 3.546),

4) Al_4TiO_8 – MgO – FeO – Mg_2TiO_4 (volume – 359 ‰, degree of asymmetry – 2.000),

5) Al_4TiO_8 – FeO – Mg_2TiO_4 – Fe_2TiO_4 (volume – 128 ‰, degree of asymmetry – 2.501),

6) Al_4TiO_8 – Mg_2TiO_4 – Fe_2TiO_4 – $FeTiO_3$ (volume – 61 ‰, degree of asymmetry – 4.047),

7) Al_4TiO_8 – Mg_2TiO_4 – $FeTiO_3$ – $MgTiO_3$ (volume – 57 ‰, degree of asymmetry – 3.820),

8) Al_4TiO_8 – $FeTiO_3$ – $MgTiO_3$ – $MgTi_2O_5$ (volume – 45 ‰, degree of asymmetry – 4.827),

9) Al_4TiO_8 – $FeTiO_3$ – $MgTi_2O_5$ – $FeTi_2O_5$ (volume – 24 ‰, degree of asymmetry – 3.915),

10) Al_4TiO_8 – $MgTi_2O_5$ – $FeTi_2O_5$ – Al_2TiO_5 (volume – 10 ‰, degree of asymmetry – 4.063),

11) $MgTi_2O_5$ – $FeTi_2O_5$ – Al_2TiO_5 – TiO_2 (volume – 35 ‰, degree of asymmetry – 2.800).

The calculated data obtained above a temperature of 2076 K (Fig. 12 and Table 6), in view of the fact that the existence of the Al_4TiO_8 compound has not been proven,[15] are of a recommendatory nature and require further theoretical and practical studies.

Analyzing all the results obtained, it should be stressed that above the temperature of 1141 K, four elementary tetrahedrons remain unchanged: MgO – FeO – Mg_2TiO_4 – $MgAl_2O_4$, $FeAl_2O_4$ – Mg_2TiO_4 – FeO – Fe_2TiO_4, $FeAl_2O_4$ – Mg_2TiO_4 – $MgAl_2O_4$ – FeO and $FeAl_2O_4$ – $MgTiO_3$ – $MgAl_2O_4$ – Al_2O_3 which have a relatively large volume. These areas have a higher reliability in predicting the phase composition of materials without special technological methods for obtaining spinel-containing materials.

Other areas can also be used to obtain various composite materials based on compositions of the MgO – Al_2O_3 – FeO – TiO_2 system. However, it is necessary to carry out additional calculations of the permissible error in the dosage of the components. Also, when synthesizing materials in areas that have a small volume and a high degree of asymmetry, special precision in the dosage of raw materials and various special technological methods are required to achieve a homogeneous distribution of components in the synthesis batch.

Table 6. Geometric-topological characteristics of the phases of the MgO – Al_2O_3 – FeO – TiO_2 system above 2076 K

Phase	Coexisting phases and lengths of conodes, L, ‰	Number of elementary tetrahedra, where the phase exists	Total volume of existence, ‰	Probability of existence, ω, fractions of units
MgO	FeO (1000), Al_4TiO_8 (893), $MgAl_2O_4$ (719), Mg_2TiO_4 (500)	2	562	0.1405
Al_2O_3	$FeAl_2O_4$ (414), Al_4TiO_8 (282), $MgAl_2O_4$ (281)	1	32	0.0080
FeO	MgO (1000), $MgAl_2O_4$ (893), Al_4TiO_8 (893), Mg_2TiO_4 (866), $FeAl_2O_4$ (586), Fe_2TiO_4 (357)	4	736	0.1840
TiO_2	Al_2TiO_5 (560), $FeTi_2O_5$ (310), $MgTi_2O_5$ (200)	1	35	0.0087
$MgAl_2O_4$	FeO (893), MgO (719), $FeAl_2O_4$ (366), Al_4TiO_8 (282), Al_2O_3 (281),	3	281	0.0702
Al_4TiO_8	FeO (893), MgO (893), Fe_2TiO_4 (684), $MgTi_2O_5$ (642), Mg_2TiO_4 (638), $FeTiO_3$ (632), $FeTi_2O_5$ (624), $MgTiO_3$ (622), $FeAl_2O_4$ (366), Al_2O_3 (282), $MgAl_2O_4$ (282), Al_2TiO_5 (158)	10	965	0.2413
Al_2TiO_5	TiO_2 (560), $MgTi_2O_5$ (492), $FeTi_2O_5$ (486), Al_4TiO_8 (158)	2	45	0.0113
$FeAl_2O_4$	FeO (586), Al_2O_3 (414), $MgAl_2O_4$ (366), Al_4TiO_8 (366)	2	78	0.0195
Fe_2TiO_4	Al_4TiO_8 (684), Mg_2TiO_4 (585), FeO (357), $FeTiO_3$ (169)	2	189	0.0472
$FeTiO_3$	Al_4TiO_8 (632), Mg_2TiO_4 (488), $MgTiO_3$ (422), $MgTi_2O_5$ (412),Fe_2TiO_4 (169), $FeTi_2O_5$ (164)	4	187	0.0467
$FeTi_2O_5$	Al_4TiO_8 (624), Al_2TiO_5 (486), TiO_2 (310), $MgTi_2O_5$ (272), $FeTiO_3$ (164)	3	69	0.0173
$MgTi_2O_5$	Al_4TiO_8 (642), Al_2TiO_5 (492), $FeTiO_3$ (412), $FeTi_2O_5$ (272), TiO_2 (200), $MgTiO_3$ (133)	4	114	0.0285
$MgTiO_3$	Al_4TiO_8 (622), $FeTiO_3$ (422), Mg_2TiO_4 (167), $MgTi_2O_5$ (133)	2	102	0.0255
Mg_2TiO_4	FeO (866), Al_4TiO_8 (638), Fe_2TiO_4 (585), MgO (500), $FeTiO_3$ (488), $MgTiO_3$ (167)	4	605	0.1513
∑			4000	1.0000

3. Conclusions

The subsolidus structure of the four-component system $MgO - Al_2O_3 - FeO - TiO_2$ was studied in six temperature ranges. Geometric-topological characteristics of the phases of the system have been determined, topological graphs of the relationship of elementary tetrahedrons have been constructed, their volumes, degrees of asymmetry for all temperature ranges have been found. A comparative analysis of the data obtained made it possible to identify the most technological areas of compositions for the production of spinel-containing materials, which are within the elementary tetrahedra: $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$, $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$, $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$ and $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$.

The results of studying the structure of the $MgO - Al_2O_3 - FeO - TiO_2$ system will also be useful to make new composite materials with a given phase composition and operational properties.

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СУБСОЛІДУСНА БУДОВА СИСТЕМИ $MgO - Al_2O_3 - FeO - TiO_2$

***Анотація**. Досліджено субсолідусну будову чотирикомпонентної системи $MgO - Al_2O_3 - FeO - TiO_2$ в шести температурних інтервалах. Визначено геометро-топологічні характеристики фаз досліджуваної системи, побудовано топологічні графи взаємозв'язку елементарних тетраедрів, визначено їхні об'єми, ступінь асиметрії для всіх температурних інтервалів. Прогнозовано оптимальні області складів для виробництва матеріалів, що містять шпінель, які лежать у межах елементарних тетраедрів: $MgO - FeO - Mg_2TiO_4 - MgAl_2O_4$, $FeAl_2O_4 - Mg_2TiO_4 - FeO - Fe_2TiO_4$, $FeAl_2O_4 - Mg_2TiO_4 - MgAl_2O_4 - FeO$ та $FeAl_2O_4 - MgTiO_3 - MgAl_2O_4 - Al_2O_3$.*

***Ключові слова:** субсолідусна будова, тетраедрація, геометро-топологічні характеристики, алюмомагнезіальна шпінель, герцинiт, кванділіт.*