STUDYING OF THE STRUCTURE OF AL₂O₃–SiO₂–CaO–P₂O₅ SYSTEM AND ITS SIGNIFICANCE IN THE TECHNOLOGY OF REFRACTORIES

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Abstract
The paper shows that presence of such phases as mullite, corundum is required in order to obtain high quality refractories that are able to work effectively under the conditions of the simultaneous effects of corrosive environments, high temperatures and pressure, sudden changes in temperature. The structures of Al₂O₃–SiO₂–CaO–P₂O₅ system are examined in the materials in which the formation of defined phases is probably. Based on data it is carried out partitioning of the system on the elementary tetrahedrons. The data on the lengths of tie lines, volumes, the asymmetry degree and the eutectic temperature of elementary tetrahedrons are given. The geometric-topological characteristic of the phases of this system are presented. The choice of oxides compositions areas for the production of refractories is justified based on the obtained results.

Keywords: geometric-topological characteristics, mullite, corundum, phase, refractories, elementary tetrahedrons, system.

DOI: 10.21303/2461-4262.2017.00367 © Yulia Kharybina, Yaroslav Pitak

1. Introduction
Systems comprising refractory oxides, and compounds as well as phosphates of the composition are of great interest for the study of the kinetics and mechanism for determining the hardening of phosphate tangles composed refractory like at normal temperature or under heating. It is important because the processes of hardening and the products of formation occur in different ways [1–4]. One of such system is the Al₂O₃–SiO₂–CaO–P₂O₅ system. This system has practical value in the technology of refractory non-metallic materials. Especially it is necessary in considering the life of refractories in thermal units of construction industry [5–7]. The first version of the partition of this system into elementary tetrahedral is given [5].

The aim of research is determination of the geometric-topological characteristics of the phases of the system, taking into account new data on the coexistence of phases and structure of the system.

According to the classification [1], Al₂O₃–SiO₂–CaO–P₂O₅ system is the system of high complexity (43 elementary tetrahedrons, 32 compounds). Absence of the thermodynamic data about this system will not allow to clarify the processes of phase formation that occur in the material at high temperatures, as well as to solve important problems associated with the scientifically grounded choice of compositions and development of rational technological methods. The study of this system will be continued due to its great value for the construction and metallurgical areas. Also it is necessary to be studied by the authors in a form such as described in [8–10].

2. Materials and Methods
The geometric-topological characteristics of phase of the system consist of: number of the tetrahedrons where this phase is in, number of phases with which coexist, the volume of existence (ΣV, the total volume of all elementary tetrahedrons where this phase is in), probability of existence (ω).

The calculating formula of probability existence of phases in this concentration tetrahedron is presented (1):
\[ \omega_i = \frac{\sum V_i}{n \times V_0}, \]  
(1)

where \( \sum V_i \) – total volume of elementary tetrahedrons in which there is this phase, \( V_0 \) – the volume of concentration tetrahedron, \( n \) – the number of components in the system, in this case \( n = 4 \).

The relative volume of the elementary tetrahedron is calculated using the determinant by the formula (2):

\[ V_i = \begin{vmatrix} X_1 & Y_1 & Z_1 & 1 \\ X_2 & Y_2 & Z_2 & 1 \\ X_3 & Y_3 & Z_3 & 1 \\ X_4 & Y_4 & Z_4 & 1 \end{vmatrix}, \]

(2)

where \( X_i, Y_i, Z_i \) – content of oxides \( \text{Al}_2\text{O}_3, \text{SiO}_2, \text{CaO}, \text{P}_2\text{O}_5 \) in the compounds constituting the elementary tetrahedron.

The degree of asymmetry of elementary tetrahedrons are estimated as the ratio of the maximum (\( L_{\text{max}} \)) to a minimum edge length (\( L_{\text{min}} \)) by the formula (3):

\[ K = \frac{L_{\text{max}}}{L_{\text{min}}}. \]

(3)

The conode length of elementary tetrahedrons is calculated using the barycentric coordinates and elements of Euclidean geometry by the formula (4):

\[
L^2 = (x_2 - x_1)^2 + (y_2 - y_1)^2 + (z_2 - z_1)^2 + \\
+ (T_2 - T_1)^2 + (x_2 - x_1)(y_2 - y_1) + (x_2 - x_1)(z_2 - z_1) + \\
+ (x_2 - x_1)(T_2 - T_1) + (y_2 - y_1)(z_2 - z_1) + \\
+ (y_2 - y_1)(T_2 - T_1) + (z_2 - z_1)(T_2 - T_1),
\]

(4)

where \( x_1, y_1, z_1, x_2, y_2, z_2 \) – the coordinates (component concentration) of coexisting vapor phase.

To illustrate the relationship of elementary tetrahedrons of system it is necessary to use the method of topological graphs, as described in [11]. The number of edges (\( R \)) is calculated according to Euler’s formula (5):

\[ R = \frac{Z_1 + 2Z_2 + 3Z_3 + 4Z_4}{2}. \]

(5)

Taking into account that the eutectic temperature of the liquid curves for all components of the system are equal, the calculation of the temperature and eutectic composition for the four-systems are produced by the decision of the system of equations (6) given in [11]:

\[
\begin{align*}
T_1 &= \frac{T_{n11}}{\ln(X_1)} = T_2 = T_{n12} / \left(1 - \frac{\ln(X_2)}{N_2}\right), \\
T_2 &= \frac{T_{n22}}{\ln(X_2)} = T_3 = T_{n23} / \left(1 - \frac{\ln(X_3)}{N_3}\right), \\
T_3 &= \frac{T_{n33}}{\ln(X_3)} = T_4 = T_{n34} / \left(1 - \frac{\ln(X_4)}{N_4}\right), \\
X_1 + X_2 + X_3 + X_4 &= 1.
\end{align*}
\]

(6)
Calculation of geometric-topological characteristics of the phases of the system is carried out using programs developed at the department of technology of ceramics, refractories, glass and enamels of NTU “KPI”.

3. Research results of the $\text{Al}_2\text{O}_3$–$\text{SiO}_2$–$\text{CaO}$–$\text{P}_2\text{O}_5$ system

To analyze the probability of formation reactions of the crystalline phases, the calculation of the free Gibbs energy are made from the equations given by [12, 13].

It is established the possibility of the occurrence of conjugate reactions by considering the structure of the $\text{Al}_2\text{O}_3$–$\text{SiO}_2$–$\text{CaO}$–$\text{P}_2\text{O}_5$ system:

1) $\text{AP} + 3\text{C}_3\text{P} + 2\text{S} \rightarrow 3\text{C}_3\text{P} + \text{A}_2\text{P}$;
2) $\text{AP} + \text{A}_3\text{S}_2 + \text{C}_3\text{P} \rightarrow \text{C}_3\text{APS}_2 + \text{A}_3\text{P}$;
3) $\text{S} + \text{C}_3\text{P} + \text{CAS}_2 \rightarrow \text{C}_3\text{APS}_2 + \text{CS}$;
4) $2\text{C}_3\text{P} + 3\text{S} + \text{C}_3\text{AS} \rightarrow \text{CAS}_2 + 2\text{C}_3\text{SP}$;
5) $\text{C}_3\text{P} + \text{CA} + \text{C}_3\text{AS} \rightarrow \text{CA}_2 + \text{C}_3\text{SP}$;
6) $\text{A}_3\text{S}_2 + \text{C}_3\text{P} \rightarrow \text{C}_3\text{APS}_2 + 2\text{A}$.

The temperature dependence of the free Gibbs energy for which is determined (Fig. 1).

Thermodynamic analysis of the reactions in $\text{Al}_2\text{O}_3$–$\text{SiO}_2$–$\text{CaO}$–$\text{P}_2\text{O}_5$ system (accepted conventions of $\text{Al}_2\text{O}_3$–$\text{A}$, $\text{SiO}_2$–$\text{S}$, $\text{CaO}$–$\text{C}$, $\text{P}_2\text{O}_5$–$\text{P}$) has allowed to establish the following co-existing phases of the pair (conodes taking place in three-dimensional space):

- $\text{C}_3\text{P}$–$\text{CAS}_2$;
- $\text{C}_3\text{APS}_2$–$\text{C}_3\text{P}$;
- $\text{C}_3\text{APS}_2$–$\text{C}_3\text{P}$–$\text{S}$–$\text{C}_3\text{APS}_2$;
- $\text{A}_3\text{S}_2$–$\text{C}_3\text{APS}_2$;
- $\text{A}_3\text{P}$–$\text{C}_3\text{APS}_2$;
- $\text{C}_3\text{APS}_2$–$\text{C}_3\text{AS}$;
- $\text{A}_3\text{S}_2$–$\text{C}_3\text{AS}$;
- $\text{C}_3\text{APS}_2$–$\text{C}_3\text{SP}$;
- $\text{C}_3\text{AS}$–$\text{C}_3\text{SP}$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{SP}$–$\text{CAS}_2$;
- $\text{C}_3\text{APS}_2$–$\text{C}_3\text{SP}$;
- $\text{C}_3\text{SP}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{SP}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$;
- $\text{C}_3\text{PS}$–$\text{CAS}_2$.

32 phases, 4 oxides constituting the system, 23 according to the number of binary oxides of simple compounds, 4 ternary compounds, 1 four-component are taken in determining the structure of the system. The system is partitioned on 45 elementary tetrahedrons in the subsolidus. The characteristics (data volume $V_i$, the temperature $T_i$ of occurrence and degree of asymmetry of the melt) are shown in Table 1.

It is evident (from Table 1 data) that the minimum temperature of the melt in the appearance of $\text{Al}_2\text{O}_3$–$\text{SiO}_2$–$\text{CaO}$–$\text{P}_2\text{O}_5$ system is equal to 854 K, and corresponding composition is located in elementary tetrahedron No. 7 ($\text{AP}$–$\text{C}_3\text{SP}$–$\text{S}$–$\text{S}$–$\text{C}_3\text{PS}$). The maximum temperature is equal to 1781 K ($\text{C}_3\text{SP}$–$\text{C}_3\text{AS}$–$\text{C}_3\text{AP}$–$\text{C}_3\text{SP}$–$\text{CAS}_2$) at which the solid phase still persists in the system.

The tetrahedrons $\text{AP}$–$\text{A}_3\text{S}_2$–$\text{S}$–$\text{C}_3\text{APS}_2$ ($V_i=133,95\text{~‰}$, $T_i=1552\text{~K}$), $\text{A}_3\text{S}_2$–$\text{C}_3\text{P}$–$\text{A}_3\text{P}$ ($V_i=49,22\text{~‰}$, $T_i=1742\text{~K}$), $\text{C}_3\text{APS}_2$–$\text{A}_3\text{S}_2$–$\text{S}$–$\text{S}$–$\text{C}_3\text{PS}$ ($V_i=39,27\text{~‰}$, $T_i=1573\text{~K}$) are the most technologically considering the volume of the elementary tetrahedron, the degree of asymmetry and minimal occurrence of melt temperature of the composition. The compositions of elementary tetrahedrons including mullite, corundum are the most appropriate for the technology of refractory materials (Table 1, tetrahedron number 15).
### Table 1
Elementary tetrahedrons of $\text{Al}_2\text{O}_3$–$\text{SiO}_2$–$\text{CaO}$–$\text{P}_2\text{O}_5$ system

<table>
<thead>
<tr>
<th>#</th>
<th>Elementary tetrahedrons</th>
<th>The degree of asymmetry</th>
<th>$T_{\text{v}}, \text{K}$</th>
<th>$V_{\text{v}}{%}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\text{AP}_3\text{CP}_2\text{SP}–\text{P}$</td>
<td>1,8</td>
<td>858</td>
<td>9,64</td>
</tr>
<tr>
<td>2</td>
<td>$\text{AP}_3\text{CP}_3\text{SP}–\text{CP}_2$</td>
<td>5,99</td>
<td>1020</td>
<td>2,54</td>
</tr>
<tr>
<td>3</td>
<td>$\text{AP}_3\text{CP}–\text{SP}–\text{C}_2\text{P}_5$</td>
<td>3,87</td>
<td>1017</td>
<td>4,36</td>
</tr>
<tr>
<td>4</td>
<td>$\text{AP}_3\text{CP}–\text{SP}–\text{A}_2\text{P}_3$</td>
<td>2,38</td>
<td>no data</td>
<td>11,16</td>
</tr>
<tr>
<td>5</td>
<td>$\text{AP}_3\text{CP}–\text{SP}–\text{A}_2\text{P}_3$</td>
<td>4,01</td>
<td>no data</td>
<td>8,02</td>
</tr>
<tr>
<td>6</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>1,90</td>
<td>855</td>
<td>39,37</td>
</tr>
<tr>
<td>7</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>2,42</td>
<td>854</td>
<td>45,01</td>
</tr>
<tr>
<td>8</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>12,23</td>
<td>1208</td>
<td>30,95</td>
</tr>
<tr>
<td>9</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>10,33</td>
<td>1251</td>
<td>36,13</td>
</tr>
<tr>
<td>10</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>4,01</td>
<td>no data</td>
<td>11,16</td>
</tr>
<tr>
<td>11</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>4,01</td>
<td>no data</td>
<td>8,02</td>
</tr>
<tr>
<td>12</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>2,38</td>
<td>no data</td>
<td>11,16</td>
</tr>
<tr>
<td>13</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>4,01</td>
<td>no data</td>
<td>8,02</td>
</tr>
<tr>
<td>14</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>2,38</td>
<td>no data</td>
<td>11,16</td>
</tr>
<tr>
<td>15</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>4,01</td>
<td>no data</td>
<td>8,02</td>
</tr>
<tr>
<td>16</td>
<td>$\text{AP}–\text{CP}–\text{SP}–\text{P}_3$</td>
<td>2,38</td>
<td>no data</td>
<td>11,16</td>
</tr>
</tbody>
</table>

The total volume 1000
The tie lines rearrangement in the ternary subsystem Al$_2$O$_3$–SiO$_2$–CaO–P$_2$O$_5$ is shown in Fig. 2.

The geometric-topological characteristics of phases of the system are presented (Table 2) [14–17].

Table 2
Geometric-topological characteristics of phases of Al$_2$O$_3$–SiO$_2$–CaO–P$_2$O$_5$ system

<table>
<thead>
<tr>
<th>No.</th>
<th>Phase</th>
<th>Number of the tetrahedrons where this phase is in</th>
<th>Number of phases with which coexist</th>
<th>The volume of existence, $\sum V_i$, %</th>
<th>Probability of the existence, $\omega_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>C</td>
<td>1</td>
<td>3</td>
<td>39,05</td>
<td>0,00976</td>
</tr>
<tr>
<td>2</td>
<td>A</td>
<td>3</td>
<td>5</td>
<td>92,54</td>
<td>0,0231</td>
</tr>
<tr>
<td>3</td>
<td>S</td>
<td>9</td>
<td>10</td>
<td>466,28</td>
<td>0,116</td>
</tr>
<tr>
<td>4</td>
<td>P</td>
<td>1</td>
<td>3</td>
<td>9,65</td>
<td>0,00241</td>
</tr>
<tr>
<td>5</td>
<td>CA</td>
<td>8</td>
<td>8</td>
<td>83,48</td>
<td>0,0208</td>
</tr>
<tr>
<td>6</td>
<td>CaA</td>
<td>6</td>
<td>6</td>
<td>43,42</td>
<td>0,0109</td>
</tr>
<tr>
<td>7</td>
<td>CA</td>
<td>6</td>
<td>7</td>
<td>49,20</td>
<td>0,0123</td>
</tr>
<tr>
<td>8</td>
<td>CA$_2$</td>
<td>2</td>
<td>4</td>
<td>27,58</td>
<td>0,00690</td>
</tr>
<tr>
<td>9</td>
<td>CA$_3$</td>
<td>3</td>
<td>5</td>
<td>83,49</td>
<td>0,0208</td>
</tr>
<tr>
<td>10</td>
<td>CS</td>
<td>3</td>
<td>5</td>
<td>56,20</td>
<td>0,0141</td>
</tr>
<tr>
<td>11</td>
<td>C$_2$S</td>
<td>5</td>
<td>7</td>
<td>43,42</td>
<td>0,0109</td>
</tr>
<tr>
<td>12</td>
<td>C$_2$S$_2$</td>
<td>2</td>
<td>4</td>
<td>13,79</td>
<td>0,00345</td>
</tr>
<tr>
<td>13</td>
<td>CS</td>
<td>7</td>
<td>8</td>
<td>154,98</td>
<td>0,0387</td>
</tr>
<tr>
<td>14</td>
<td>C$_3$P</td>
<td>4</td>
<td>6</td>
<td>54,07</td>
<td>0,0135</td>
</tr>
<tr>
<td>15</td>
<td>C$_3$P$_5$</td>
<td>18</td>
<td>17</td>
<td>378,97</td>
<td>0,0948</td>
</tr>
<tr>
<td>16</td>
<td>C$_3$P$_5$</td>
<td>4</td>
<td>5</td>
<td>140,40</td>
<td>0,0351</td>
</tr>
<tr>
<td>17</td>
<td>C$_3$P$_5$</td>
<td>2</td>
<td>4</td>
<td>67,06</td>
<td>0,0168</td>
</tr>
<tr>
<td>18</td>
<td>C$_5$P$_5$</td>
<td>6</td>
<td>8</td>
<td>138,82</td>
<td>0,0347</td>
</tr>
<tr>
<td>19</td>
<td>C$_5$P$_5$</td>
<td>2</td>
<td>4</td>
<td>6,89</td>
<td>0,00173</td>
</tr>
<tr>
<td>20</td>
<td>C$_5$P$_5$</td>
<td>2</td>
<td>4</td>
<td>12,18</td>
<td>0,00305</td>
</tr>
<tr>
<td>21</td>
<td>A$_3$S$_2$</td>
<td>7</td>
<td>7</td>
<td>323,99</td>
<td>0,0810</td>
</tr>
<tr>
<td>22</td>
<td>A$_3$P</td>
<td>3</td>
<td>5</td>
<td>114,21</td>
<td>0,0286</td>
</tr>
<tr>
<td>23</td>
<td>AP</td>
<td>10</td>
<td>11</td>
<td>442,94</td>
<td>0,111</td>
</tr>
<tr>
<td>24</td>
<td>A$_3$P$_3$</td>
<td>2</td>
<td>4</td>
<td>19,18</td>
<td>0,00480</td>
</tr>
<tr>
<td>25</td>
<td>AP$_5$</td>
<td>4</td>
<td>6</td>
<td>27,69</td>
<td>0,00692</td>
</tr>
<tr>
<td>26</td>
<td>S$_3$P</td>
<td>2</td>
<td>4</td>
<td>84,35</td>
<td>0,0211</td>
</tr>
<tr>
<td>27</td>
<td>SP</td>
<td>6</td>
<td>8</td>
<td>75,05</td>
<td>0,0188</td>
</tr>
<tr>
<td>28</td>
<td>C$_3$AS</td>
<td>11</td>
<td>10</td>
<td>167,20</td>
<td>0,0418</td>
</tr>
<tr>
<td>29</td>
<td>CAS$_5$</td>
<td>8</td>
<td>8</td>
<td>243,38</td>
<td>0,0608</td>
</tr>
<tr>
<td>30</td>
<td>C$_3$S$_5$P</td>
<td>12</td>
<td>10</td>
<td>79,12</td>
<td>0,0198</td>
</tr>
<tr>
<td>31</td>
<td>C$_3$SP</td>
<td>10</td>
<td>8</td>
<td>54,30</td>
<td>0,0136</td>
</tr>
<tr>
<td>32</td>
<td>C$_3$APS$_5$</td>
<td>10</td>
<td>8</td>
<td>407,12</td>
<td>0,102</td>
</tr>
<tr>
<td>33</td>
<td>The total volume</td>
<td>179</td>
<td>212</td>
<td>4000,00</td>
<td>1,0000</td>
</tr>
</tbody>
</table>

C$_3$P, AP, C$_3$SP, C$_3$AS, S phases (respectively – No. 15, 23, 31, 28, 3) have the largest number of coexisting phases (Table 2). C$_3$P phase presents in 18 elementary tetrahedron and has a maxi-
mum amount of existence – 424.6 ‰. Significant volumes of existence in this system have a phase:
S (481.5), AP (414.2), C\textsubscript{3}APS\textsubscript{2} (396.0), A\textsubscript{3}S (287.5).

Fig. 2. The state of elementary tetrahedrons of Al\textsubscript{2}O\textsubscript{3}–SiO\textsubscript{2}–CaO–P\textsubscript{2}O\textsubscript{5} system in the concentration tetrahedron

4. Discussing of research results
Comparative analysis of the data is revealed the most technological range of compositions for the production of refractory products. They are located in the immediate vicinity to the edges of elementary tetrahedrons AP–A\textsubscript{3}S\textsubscript{2}–S–C\textsubscript{3}APS\textsubscript{2}; A–A\textsubscript{3}S\textsubscript{2}–C\textsubscript{3}P–A\textsubscript{3}P; C\textsubscript{3}APS\textsubscript{2}–A\textsubscript{3}S\textsubscript{2}–S–CAS\textsubscript{2}.

To the basis of the structure of the system data it is of interest to modeling changes the phase structure in the compositions of mullite refractory – slag in the future. It will allow coming nearly to explain the reasons for the destruction of refractories in service when the ratio of the components and their interaction temperature are changing [18].

5. Conclusions
The geometric-topological characteristics of the phases of Al\textsubscript{2}O\textsubscript{3}–SiO\textsubscript{2}–CaO–P\textsubscript{2}O\textsubscript{5} system are defined; there are identified the elementary volume, the degree of asymmetry and the eutectic temperature of the elementary tetrahedrons.
The results of studies on the structure of the four-component system (Al\textsubscript{2}O\textsubscript{3}–SiO\textsubscript{2}–CaO–P\textsubscript{2}O\textsubscript{5}) serve as a theoretical basis for further developments in the field of new technology of refractory non-metallic materials.

References


