Thermochemical study of natural pollucite

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Abstract

Experimental data on the heat capacities, enthalpies of formation from the oxides and elements of the natural pollucites: $Cs_{0.77}Na_{0.14}Rb_{0.04}Al_{0.91}Si_{2.08}O_6 \cdot 0.34H_2O$ (Minas Gerais, Brazil) (I) and $Cs_{0.84}Na_{0.11}Al_{0.88}Si_{2.10}O_6 \cdot 0.17H_2O$ (East Siberia, Russia) (II) have been determined. The heat capacities of the pollucite (II) were measured by differential scanning calorimetry (DSC) method from $T = 250$ to 810 K. A value of $C^\circ_{p,m}(298.15\,\text{K}) = 172.2\,\text{J K}^{-1}\,\text{mol}^{-1}$ was obtained and the resultant $C^\circ_{p,m}(T)$ equation in the interval $T = 298.15\text{--}610$ K was calculated: $C^\circ_{p,m} = 131.37 + 181.97 \times 10^{-3}T - 11.84 \times 10^5 T^{-2}\,\text{J K}^{-1}\,\text{mol}^{-1}$ ($\pm 0.36\%$). The standard enthalpies of formation were determined by a set of thermochemical cycles using decomposition and subsequent dissolution of the dehydration products in molten lead borate at $T = 973$ K. Values of $\Delta_f H^\circ_m(298.15\,\text{K})$ are found to be $-3104 \pm 13\,\text{kJ mol}^{-1}$ (pollucite I) and $-3090 \pm 14\,\text{kJ mol}^{-1}$ (pollucite II). The values of $\Delta_f G^\circ_m(298.15\,\text{K})$ were calculated as $-2921\,\text{kJ mol}^{-1}$ (pollucite I) and $-2911\,\text{kJ mol}^{-1}$ (pollucite II).
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1. Introduction

Pollucite is found in rare-element-rich granitic pegmatites having lithium and niobium–tantalum mineralization. It is an aluminosilicate phase with a cubic lattice representing the Cs end-member of the analcime $NaAlSi_2O_6 \cdot H_2O$–leucite $KAlSi_2O_6$–pollucite $CsAlSi_2O_6$ series. Replacement of Cs ions by Na, Rb and Li occurs and mixing with Na is more significant than with alkaline elements. It may be connected with the fact that pollucite and analcime are isostructural ($a = 13.67 \times 10^{-10}$ m for natural pollucite and

$a = 13.66 \times 10^{-10}$ m for analcime). The typical compositions of natural pollucite can be described by $Cs_{1-x}(Na, K, Rb, Li)_x AlSi_2O_6 \cdot xH_2O$ ($0 \leq x \leq 1$). Sodium-poor pollucite close to the end-member composition $CsAlSi_2O_6$ has been described only on a microscopic scale [1]. Pollucite abundances in pegmatites can be economically significant and serve as a source of cesium. Synthetic pollucite is an important long-term containment phase of cesium radioisotopes. Analcime–pollucite phases commonly crystallize at low temperatures during hydrothermal alteration of nuclear waste forms [2,3].

The knowledge of the thermodynamic properties of pollucite, analcime, leucite and their solid solutions is of geological and technological interest. We have determined previously the thermodynamic properties of

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Table 1
The thermodynamic properties pollucite at $T = 298.15$ K

Mineral	S_m° (J K^{-1} mol^{-1})	$-\Delta_f H_m^\circ$ (kJ mol^{-1})	$-\Delta_f G_m^\circ$ (kJ mol^{-1})	Reference
$Cs_{0.65}Na_{0.185}Rb_{0.028}AlSi_2O_{5.863}(OH)_{0.132} \cdot 0.19H_2O$ (molar mass = 275.66 $\times$ 10^{-3} kg mol^{-1})	207.2 $\pm$ 0.4	3098.5 $\pm$ 3.6	2921.6 $\pm$ 3.6	[6]
$CsAlSi_2O_6$ (synthetic) (molar mass = 312.055 $\times$ 10^{-3} kg mol^{-1})	208.2 $\pm$ 0.6	3083.4 $\pm$ 5.0	2917	[7]
$Cs_{0.77}Na_{0.14}Rb_{0.04}Al_{0.91}Si_{2.08}O_6 \cdot 0.34H_2O$ (molar mass = 294.15 $\times$ 10^{-3} kg mol^{-1})	221	3104 $\pm$ 13	2921[a]	This work
$Cs_{0.84}Na_{0.11}Al_{0.88}Si_{2.10}O_6 \cdot 0.17H_2O$ (molar mass = 295.88 $\times$ 10^{-3} kg mol^{-1})	197	3090 $\pm$ 14	2911[a]	This work

[a] Values are calculated using the S_m°(298.15 K) from [6].

analcime [4] and leucite [5]. This paper is concerned with an experimental investigation of the thermodynamic properties of pollucite. The thermodynamic properties of a natural pollucite with the composition $Cs_{0.65}Na_{0.185}Rb_{0.028}AlSi_2O_{5.863}(OH)_{0.132} \cdot 0.19H_2O$ were investigated by Bennington et al. [6] (Table 1) using different calorimetric methods: acid (HF) solution calorimetry to obtain the enthalpy of formation, "drop" calorimetry method to determine the heat content and adiabatic calorimetry to measure the low-temperature heat capacities and to calculate the value of the standard entropy [7] (Table 1) studied two samples of synthetic anhydrous pollucite. The enthalpy of formation was determined by high-temperature solution calorimetry for a sample $Cs_{0.90}Al_{1.01}Si_{2.06}O_6$, which was synthesized by hydrothermal methods. The heat capacities of the same sample and a sample of composition $Cs_{0.90}Al_{1.09}Si_{2.00}O_{6.00}$ synthesized by solid-state reaction were measured by adiabatic calorimetry. Semenov et al. [7] calculated the thermodynamic properties for the composition of anhydrous pollucite $CsAlSi_2O_6$. The values of thermodynamic data obtained by Bennington et al. [6] and Semenov et al. [7] do not agree with one another as they correspond to different compositions. Bennington et al.'s [6] data on the entropy and $C_p(T)$ equation for natural pollucite $Cs_{0.65}Na_{0.185}Rb_{0.028}AlSi_2O_{5.863}(OH)_{0.132} \cdot 0.19H_2O$ are included in the compilation of Robie and Hemingway [8], while the values for the enthalpy and Gibbs free energy of formation are absent.

The goal of this investigation is an extention of the experimental data on fundamental properties (the heat capacity, the enthalpies of formation from oxides and elements) of natural pollucite by differential scanning calorimetry (DSC) and heat-flux Calvet microcalorimetry.

2. Sample characterization

We chose for thermochemical investigation well-crystallized natural samples of pollucite. The first pollucite sample (I) is from Minas Gerais, Brazil. Transparent, and semitransparent colorless crystals up to 5×10^{-3} m in dimension were found in the vugs of granitic pegmatites with spodumene, amethyst, albite and other silicates. The pollucite sample (II) is from East Siberia, Russia. Transparent, clean, colorless cubic crystals up to $(10–20) \times 10^{-3}$ m size were separated from vugs of granitic pegmatites. Pure crystals or pieces of single crystals were separated under a microscope and samples of 2×10^{-6} kg weight were chosen.

The chemical compositions of the minerals given in Table 2 were determined by a "Camebax" electron microprobe with WDS spectrometers and an EDS system "Link 860/500". The amount of zeolite water was determined by weighing the sample before and after heating at $T = 973$ K with a precision of $\pm 2 \times 10^{-9}$ kg which gave 2.11 wt.% (sample I) and 1.01 wt.% (sample II). The formulae of the two pollucites are: $Cs_{0.77}Na_{0.14}Rb_{0.04}Al_{0.91}Si_{2.08}O_6 \cdot 0.34H_2O$ (I) and $Cs_{0.84}Na_{0.11}Al_{0.88}Si_{2.10}O_6 \cdot 0.17H_2O$ (II). Teerstra and Cerny [1] showed composition histograms of 71 pollucites reported in the literature. It was established that typical natural pollucites contain 70–85 mol% of its CRK end-member components [100(Cs + Rb + K)/sum of cations]. The Si/Al ratio is generally greater than the ideal value of 2.0, typically 2.1–2.5 [1,9]. The samples studied in this work have the CRK ratios equal to 85 and 88, Si/Al ratios 2.3 and 2.4, respectively.

Powder X-ray diffraction (XRD) measurements were carried out with DRON UM-1 diffractometer

Table 2
The chemical composition and lattice parameters of natural pollucites

	Pollucite I (Minas Gerais, Brazil)	Pollucite II (East Siberia, Russia)
Chemical composition (wt.%)		
SiO_2	42.29	42.62
Al_2O_3	15.67	15.14
Cs_2O	36.64	40.25
Na_2O	1.47	1.12
K_2O	0.04	0.03
Rb_2O	1.23	–
MgO	–	0.11
FeO	–	0.03
H_2O	2.11	1.01
Total	99.45	100.31
Cations and H_2O molecules in formulae		
Si	2.08	2.10
Al	0.91	0.88
Cs	0.77	0.84
Na	0.14	0.11
Rb	0.04	–
H_2O	0.34	0.17
Lattice parameter		
a ($\times 10^{-10}$ m)	13.6796	13.6520

using Co Kα radiation. A thin layer of the pollucite was mounted on a glass slide, and the data were collected from 26 to 68° (2Θ) with steps of 0.05° and a step time of 2 s. Metallic Si powder was used as an internal standard for determination of the unit-cell constant. The diffraction patterns showed that the samples are monophasic and they assume the *Ia*3*d* structure. The lattice parameters of the samples studied are given in Table 2. They agree well with reference data: $a = 13.674 \times 10^{-10}$ m (JCPDS # 29–194).

High-temperature behavior was studied by Derivatograph Q-1500D (Hungary) between $T = 293$ and 1273 K with heating rate of $0.25 \, K \, s^{-1}$ in the air atmosphere. The masses of the samples studied were about 100×10^{-6} ($\pm 2 \times 10^{-7}$) kg. DTA and TG curves showed small endothermic effects connected with loss of water in the temperature intervals of $T = 623$–1023 K (2.1%) for sample I and $T = 843$–1033 K (1.0%) for sample II. The difference in the dehydration temperature of samples can be explained by higher containing of analcime component, which looses its water at lower temperature [4].

3. Experimental techniques

The enthalpies of formation of pollucite were determined with a Tian–Calvet high-temperature heat-flux microcalorimeter ("Setaram") under static air atmosphere. We used this device previously to study minerals, containing water and hydroxyl groups (malachite, azurite [10], dioptase [11], and natural zeolites [12]. A thermochemical cycle, which takes advantage of the rapid decomposition was utilized to obtain the enthalpy of formation. First the sample of pollucite was dropped from room temperature ($T = 298.15$ K) into the calorimeter at $T = 973$ K with no solvent present. The heat measured contained two contributions: the heat content of the sample and the enthalpy of dehydration at $T = 973$ K ($\Delta_{298.15}^{973} H_m^\circ + \Delta_{\text{dehydration},973} H_m^\circ = \Delta_{\text{drop dehydration}} H_m^\circ$). The amount of the free water was determined by weighing the sample before and after the run with a precision of $\pm 2 \times 10^{-9}$ kg, and this indicated complete dehydration. The water-free sample was removed from calorimeter, then dropped again in a second step into the calorimeter from $T = 298.15$ to 973 K, whereby the heat content ($\Delta_{298.15}^{973} H_m^\circ) = \Delta_{\text{drop}} H_m^\circ$ was measured. Then the enthalpy of solution of dehydrated pollucite at $T = 973$ K was determined. An amount of $(10–20) \times 10^{-6}$ kg of sample was placed in a foil cup and then equilibrated thermally in the calorimeter usually 7200–10,800 s before being dissolved in about 30×10^{-3} kg of molten $2PbO \cdot B_2O_3$. Dissolution of the sample is initiated by dipping the cup with the sample into the melt solvent. Stirring is manual by moving the platinum cup up and down several times. The sample dissolved readily and gave a complete reaction within 1800–3000 s before a steady baseline signal was restored. The same method was used to measure the enthalpy of solution of corundum α-Al_2O_3 (the standard sample of thermodynamic properties). The "transposed temperature drop calorimetry" method was used to obtain the enthalpies of solution of pollucite (II) and cesium chloride ("high purity", >99.9%). In each run, $(5–10) \times 10^{-6}$ kg of sample was dropped from room temperature into molten $2PbO \cdot B_2O_3$ at $T = 973$ K. The heat effect measured was thus the sum of the heat content and the enthalpy of solution of the mineral ($\Delta_{298.15}^{973} H_m^\circ + \Delta_{\text{solution},973} H_m^\circ = \Delta_{\text{drop solution}} H_m^\circ$). The behavior of water in high-temperature lead

borate solution calorimetry of volatile-bearing phases was studied by Kiseleva and Ogorodova [13] and Navrotsky et al. [14]. They showed that the water interacts with melt and dissolves in it. Calibration of the calorimeter was performed by dropping pieces of platinum wire (in the solution experiments) and α-Al_2O_3 (in the "drop" experiments), and using their known enthalpies [8].

Heat capacity measurements were performed using a heat-flux DSC "Mettler TA-2000B". The interval between the initial and final temperatures was $T = 250$–810 K. Six scans were made for checking the experimental precision. Samples of mass $(30$–$50) \times 10^{-6}$ kg were sealed in aluminum pans and placed in calorimeter. The experiments were made under flowing nitrogen with a heating rate of 0.17 K s^{-1}. The molar melting enthalpy and melting temperature of indium (99.9999% purity) were used to calibrate the calorimeter. Reproducibility of enthalpy and temperature of melting experiments were ± 0.3 and $\pm 0.01\%$, respectively. The errors in the heat capacity measurements, based on earlier runs performed on a standard substance (fine-grained α-Al_2O_3), were ± 2–3% at temperatures lower than $T = 300$ K and ± 1.5–2% between $T = 300$ and 810 K.

4. Results and discussion

The heat capacity of pollucite (II) was measured between $T = 250$ and 810 K. The average results of six scans of heat capacity measurements are presented in Table 3. These data were fitted by least-squares methods yielding, with respective average deviation of approximation presented in parenthesis:

$$C^{\circ}_{p,m} = 131.37 + 181.97 \times 10^{-3} T$$
$$- 11.84 \times 10^{5} T^{-2} \text{ J K}^{-1} \text{ mol}^{-1} \ (\pm 0.36\%)$$
$$\text{at } 298.15\text{–}610 \text{ K.}$$

At $T = 298.15$ K, the molar heat capacity of natural pollucite $Cs_{0.84}Na_{0.11}Al_{0.88}Si_{2.10}O_6 \cdot 0.17H_2O$ is 172.2 J K^{-1} mol^{-1}. This value is in agreement with experimental adiabatic calorimetric determination on natural pollucite $Cs_{0.65}Na_{0.185}Rb_{0.028}AlSi_2O_{5.863}$ $(OH)_{0.132} \cdot 0.19H_2O$ (173.9 J K^{-1} mol^{-1}) [6] and for an anhydrous synthetic sample $CsAlSi_2O_6$ (173.2 J K^{-1} mol^{-1}) [7]. The differences connected
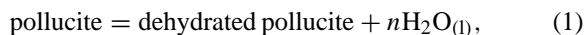
Table 3
Experimental heat capacities of pollucite (II) (molar mass $= 295.88 \times 10^{-3}$ kg mol^{-1})

T (K)	$C^{\circ}_{p,m}$ ($\times 10^3$ J K^{-1} kg^{-1})	T (K)	$C^{\circ}_{p,m}$ ($\times 10^3$ J K^{-1} kg^{-1})
280	0.569	450	0.694
290	0.577	460	0.710
300	0.585	470	0.720
310	0.593	480	0.732
320	0.601	490	0.736
330	0.610	500	0.743
340	0.619	510	0.749
350	0.627	520	0.756
360	0.636	530	0.764
370	0.641	540	0.767
380	0.646	550	0.774
390	0.654	560	0.775
400	0.660	570	0.784
410	0.668	580	0.785
420	0.676	590	0.793
430	0.684	600	0.793
440	0.687	610	0.799

with variations in chemical composition between the samples are not more than $\pm 1.0\%$.

The Calvet microcalorimetry results are given in Table 4, experimental errors are expressed at the 95% confidence level, number of determinations is given in parentheses. The enthalpies of formation of the two pollucites were determined by a set of thermochemical cycles, using decomposition and subsequent dissolution of the dehydration products. The standard enthalpy for the reaction:

$$\text{pollucite} = \text{dehydrated pollucite} + n\text{H}_2\text{O}_{(l)}, \qquad (1)$$

is calculated using experimental data (Table 4) and the equation:

$$\Delta_{\text{reaction (1)}} H^{\circ}_{m}(298.15 \text{ K})$$
$$= \Delta_{\text{drop dehydration}} H^{\circ}_{m}(\text{pollucite})$$
$$- \Delta_{\text{drop}} H^{\circ}_{m}(\text{dehydrated pollucite})$$
$$- \Delta_{\text{drop}} H^{\circ}_{m}(\text{H}_2\text{O}_{(l)}), \qquad (2)$$

where n is the number of water molecules. The values of the enthalpies of reaction (1) were found to be 8.0 ± 10.4 and 14.0 ± 4.7 kJ for the pollucite I and II samples, respectively. To determine the standard molar enthalpies of formation of pollucites we need to calculate the enthalpies of formation of the dehydrated

Table 4
Results of Calvet microcalorimeter experiments at $T = 973$ K

Mineral	Molar mass $(\times 10^{-3}$ kg mol$^{-1})$	$\Delta_{\text{drop}} H_m^\circ$ (kJ mol^{-1})	$\Delta_{\text{drop dehydration}} H_m^\circ$ (kJ mol^{-1})	$\Delta_{\text{drop solution}} H_m^\circ$ (kJ mol^{-1})	$\Delta_{\text{solution}} H_m^\circ$ (kJ mol^{-1})
Pollucite $Cs_{0.77}Na_{0.14}Rb_{0.04}Al_{0.91}$ $Si_{2.08}O_6 \cdot 0.34H_2O$	294.15		187.6 ± 8.3 (4)[a]		
Dehydrated pollucite	287.94	156.3 ± 6.2 (6)			45.6 ± 3.1 (4)
Pollucite $Cs_{0.84}Na_{0.11}Al_{0.88}Si_{2.10}O_6$ $\cdot 0.17H_2O$	295.88		156.9 ± 2.6 (9)	247.1 ± 7.4 (5)	
Dehydrated pollucite	292.89	131.4 ± 4.0 (8)			105.2 ± 12.3 (5)

[a] Errors are expressed by interval for $P = 95\%$. The number of determinations is given in parenthesis.

pollucites from the constituent oxides via:

$$
\Delta_{\text{f,ox}} H_m^\circ (298.15 \text{ K})(\text{dehydrated pollucite})
$$
$$
= \sum v_i (\Delta_{\text{drop}} H_m^\circ + \Delta_{\text{solution}} H_m^\circ) \text{ox}_i
$$
$$
- (\Delta_{\text{drop}} H_m^\circ + \Delta_{\text{solution}} H_m^\circ)
$$
$$
(\text{dehydrated pollucite}), \qquad (3)
$$

where v_i is the stoichiometric coefficients in the reaction of pollucites formation from the oxides, the values $\Delta_{\text{drop}} H_m^\circ$, $\Delta_{\text{solution}} H_m^\circ$ and $\Delta_{\text{drop solution}} H_m^\circ$ are the heat contents, heats of solution and drop solution of all constituent oxides and dehydrated pollucites under the same calorimetric conditions (Tables 4 and 5).

Because of impossibility to measure the enthalpy of solution of dicesium monoxide Cs_2O in the $2PbO \cdot B_2O_3$ melt at $T = 973$ K, we estimated this value (Table 5) from the exchange reaction:

$$
Na_2O_{(s)} + 2CsCl_{(s)} = 2NaCl_{(s)} + Cs_2O_{(s)}, \qquad (4)
$$

using experimental data (Table 5) and reference enthalpies increments and enthalpies of formation

Table 5
The calorimetric data used for calculation of enthalpies of formation of pollucite at $T = 973$ K

Substance	$\Delta_{\text{solution}} H_m^\circ$ (kJ mol^{-1})	$\Delta_{\text{drop solution}} H_m^\circ$ (kJ mol^{-1})	Reference
$CsCl_{(s)}$		72.1 ± 1.0 (7)	This work
$Cs_2O_{(s)}$		−230.0 ± 4.1	This work
$Al_2O_{3(s)}$	32.71 ± 0.59 (8)		This work
$H_2O_{(l)}$	−16.9 ± 3.1		This work
$NaCl_{(s)}$		65.2 ± 1.4	[18]
$Na_2O_{(s)}$		−111.8 ± 0.9[a]	[19]
$SiO_{2(s)}$	−4.23 ± 0.21		[20]

[a] Value was calculated from drop solution of $Na_2CO_{3(s)}$ data using new value of $\Delta_f H_m^\circ (298.15 \text{ K})$ $Na_2CO_{3(s)}$ from [8].

of all substances from Robie and Hemingway [8], Rb_2O and Cs_2O from JANAF [15]. Pollucite II contains a small quantity of dirubidium monoxide. We estimated the enthalpy of drop solution of Rb_2O (-212 ± 18 kJ mol^{-1}) by correlation between drop solution data for K_2O [4] and Cs_2O. The value of enthalpy of solution of α-Al_2O_3 (32.71 ± 0.59 kJ mol^{-1}) determined in this work is in a good agreement with our previous data (33.01 ± 1.63 kJ mol^{-1}) [16] and Charlu et al.'s results (32.34 ± 0.33 kJ mol^{-1}) [17].

The values of $\Delta_{\text{f,ox}} H_m^\circ (298.15 \text{ K})$ of the dehydrated pollucites I and II were calculated to be -172.8 ± 7.1 and -209.1 ± 13.2 kJ mol^{-1}, respectively.

By combining the values of $\Delta_{\text{reaction (1)}} H_m^\circ (298.15 \text{ K})$ and $\Delta_{\text{f,ox}} H_m^\circ (298.15 \text{ K})$ of dehydrated pollucites, we determined the values of the enthalpies of formation of natural (water-containing) minerals from oxides: -180.8 ± 12.6 and -223.2 ± 14.0 kJ mol^{-1} for samples I and II, respectively.

The standard molar enthalpies of formation from the elements were obtained using the reference thermodynamic data for the constituent oxides [8] to be: -3104 ± 13 kJ mol^{-1} (sample I) and -3090 ± 14 kJ mol^{-1} (sample II) (Table 1). The enthalpy of formation of pollucite (II) can be calculated also from its experimental drop solution data (Table 4). The needed value of the enthalpy of "interaction" of water with solvent was estimated from our calorimetric data [3] for analcime. The value of enthalpy of formation from oxides of pollucite (II), calculated with this estimated value for water was equal to -211.1 ± 7.6 kJ mol^{-1} and the corresponding standard value of $\Delta_f H_m^\circ (298.15 \text{ K})$ of natural pollucite (II) $= -3077.6 \pm 7.9$ kJ mol^{-1}. These data are

in a good agreement with our results obtained using the thermochemical cycle included dehydration. Although the $\Delta_f H_m^\circ(298.15\,\text{K}) = -3090 \pm 14\,\text{kJ}\,\text{mol}^{-1}$ was determined with more uncertainty, we adopt this as our preferred value because it does not include the estimated value of enthalpy of water solution.

We recommended the following values of $\Delta_f H_m^\circ$ $(298.15\,\text{K}) = 3104 \pm 13\,\text{kJ}\,\text{mol}^{-1}$ for natural pollucite $Cs_{0.77}Na_{0.14}Rb_{0.04}Al_{0.91}Si_{2.08}O_6 \cdot 0.34H_2O$ (I) and $\Delta_f H_m^\circ(298.15\,\text{K}) = -3090 \pm 14\,\text{kJ}\,\text{mol}^{-1}$ for natural pollucite $Cs_{0.84}Na_{0.11}Al_{0.88}Si_{2.10}O_6 \cdot 0.17H_2O$ (II).

The enthalpy of formation of Rb-containing pollucite (I) obtained in this work is in a good agreement with that determined by HF-calorimetry for a Rb-containing pollucite of similar composition [6] (Table 1). The enthalpy of formation of pollucite II is close to that of synthetic anhydrous pollucite (Table 1).

The standard entropies for pollucites were estimated from adiabatic calorimetric data [6] for natural pollucites (Table 1), taking into account their compositional variations (components were considered as constituent oxides [8]). The values of standard Gibbs energies of formation from elements of pollucites (Table 1) were calculated using the thermodynamic data of constituent elements taken from Robie and Hemingway [8] and obtained entropies of pollucites studied. The values of $\Delta_f G_m^\circ(298.15\,\text{K})$ presented in Table 1 for pollucites with different chemical composition are close to one another. Thermodynamic data on pollucite studied in this work, analcime and leucite studied earlier [3,4] are valuable to calculate the physicochemical conditions of their geological formation and possible reactions induced by the burial of radioactive materials.

Acknowledgements

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