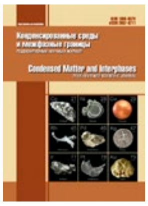

ISSN 1606-867X (Print)
ISSN 2687-0711 (Online)
Condensed Matter and Interphases
Kondensirovannye Sredy i Mezhfaznye Granitsy
https://journals.vsu.ru/kcmf/

Short communications

Short communication
https://doi.org/10.17308/kcmf.2024.26/12453

Evaluation of the thermodynamic stability of $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm) hexaaluminates with a magnetoplumbite structure in the high temperature region

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Abstract

This study is important due to the lack of reliable data about the properties of high temperature materials for energy production and aerospace engineering. The purpose of this article was to evaluate the thermodynamic stability of RE magnesium hexaaluminates $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm) with a magnetoplumbite structure, which are promising components for thermal barrier coatings. For this, we calculated the values of the Gibbs energy of the decomposition reactions of RE magnesium hexaaluminates into simple oxides and aluminum-magnesium spinel $MgAl_2O_4$ and $REAlO_3$ phases in the temperature range of 298–1,800 K. For calculations, we used data on the thermodynamic properties of hexaaluminates calculated from the values of heat capacity measured by differential scanning calorimetry in the range of 300-1,800 K and from values of thermodynamic properties of simple oxides, $MgAl_2O_4$, and $REAlO_3$ provided in previous research. There is hardly any information about the thermodynamic properties of RE magnesium hexaaluminates, which are promising thermal barrier materials. The purpose of the article is to provide a thermodynamic evaluation of the probability of decomposition reactions of hexaaluminates in the high temperature region.

Previously published data on the high temperature heat capacity of compounds with the composition of $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm) were used to calculate temperature dependences of entropy and changes in enthalpy, which were used to evaluate the Gibbs energy of the decomposition reactions of hexaaluminates into constituent oxides.

The temperature dependences of the Gibbs energy of the four possible decomposition reactions of hexaaluminates allowed drawing conclusions about thermodynamic stability in the high temperature region.

Keywords: Hexaaluminates, Magnetoplumbite, RE, Thermodynamics, Thermal barrier coatings

Funding: The study was supported by the Russian Science Foundation (grant No. 23-13-00051).

For citation: Gavrichev K. S., Guskov V. N., Gagarin P. G., Guskov A. V. Evaluation of the thermodynamic stability of $REMgAl_{11}O_{19}$ (RE=La, Pr, Nd, Sm) hexaaluminates with a magnetoplumbite structure in the high temperature region. *Condensed Matter and Interphases*. 2024;26(4): 782–788. https://doi.org/10.17308/kcmf.2024.26/12453

Для цитирования: Гавричев К. С., Гуськов В. Н., Гагарин П. Г., Гуськов А. В. Оценка термодинамической стабильности гексаалюминатов $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm) со структурой магнетоплюмбита в области высоких температур. *Конденсированные среды и межфазные границы*. 2024;26(4): 782–788. https://doi.org/10.17308/kcmf.2024.26/12453

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1. Introduction

The improved efficiency of modern power turbine plants and aircraft engines largely depends on the development of new materials that allow significantly increasing the temperature of gases in the working area. Parts from nickel-cobalt alloys used for the manufacture of critical parts (for example, turbine blades) can be effectively operated, even with cooling, at temperatures that do not exceed 1,000–1,200 °C [1]. Oxide coatings of metal parts in combination with the cooling of inner surfaces allow increasing the temperature of working gases by hundreds of degrees due to a large temperature gradient in the oxide layer [2, 3]. Coatings designed to protect against the effects of high temperature are known as thermal barrier coatings. Another important function of oxide coatings is protection against chemical exposure to substances in gaseous and condensed states, which are formed during fuel combustion and in the form of suspended particles enter the turbine together with pumping-in air [4].

Until recently, thermal barrier coatings were mainly made of yttria stabilized zirconia, YSZ [5]. This substance has some disadvantages, i.e. temperature restrictions for its application (about 1,200 °C) associated with the presence of a phase transition [6] and a significant diffusion of oxygen at high temperatures leading to the oxidation of the surfaces of metal parts. Therefore, a number of high-temperature complex RE oxides have been proposed for application: zirconates $RE_2Zr_2O_7$ [7], hafnates $RE_2Hf_2O_7$ and RE_2O_3 $2HfO_2$ [8], tantalates $RETaO_4$ and RE_3TaO_7 [9, 10], niobates RE_3NbO_7 [11], etc. These materials meet the key requirements for thermal barrier coatings: they have high melting temperatures, no phase transitions in a wide range of temperatures, have low thermal conductivity, a specified coefficient of thermal expansion, and mechanical properties. Currently, there has been a lot of interest in RE magnesium hexaaluminates [12] due to their lower thermal conductivity and potential chemical resistance to CMAS oxides (CaO, MgO, Al_2O_3, and SiO_2) at high temperatures [13].

One of the ways to evaluate if a particular oxide of thermal barrier coatings can be used under the conditions of high temperatures and the corrosive effect of gases and substances in the condensed state (in particular, melts) is the thermodynamic evaluation of the probability of decomposition reactions of complex oxides into more simple oxides, as well as reactions of interaction with the substances in the environment in the high temperature region. For this, it is necessary to determine the Gibbs energy of these reactions.

The triple phase diagram of RE_2O_3-MgO-Al_2O_3 published in [14] is characterized by the presence of 4 eutectics and a number of phases (La_2O_3, MgO, Al_2O_3, MgO·Al_2O_3, La_2O_3·Al_2O_3, $2La_2O_3$·$11Al_2O_3$) (Fig. 1). It can be noted that it does not have the $LaMgAl_{11}O_{19}$ phase with a magnetoblumbite structure. It can be assumed that in addition to $REAlO_3$ perovskites, the quasibinary diagram of RE_2O_3-Al_2O_3 for other rare-earth elements, starting with terbium, will have other compounds: aluminum garnets $RE_3Al_5O_{12}$ and $RE_4Al_2O_9$ with a monoclinic structure.

There is little information about the experimental determination of the thermodynamic properties of hexaaluminates with a magnetoplumbite structure. For example, such data are only available for heat capacity. In [15], the heat capacity of $LaMgAl_{11}O_{19}$ was determined by means of thermoanalytical analysis. The resulting data was presented in the form of a small graph. In [16], to determine the thermal conductivity of $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm, Eu, Gd), the authors used the values of specific heat capacity which were calculated by the Neumann–Kopp

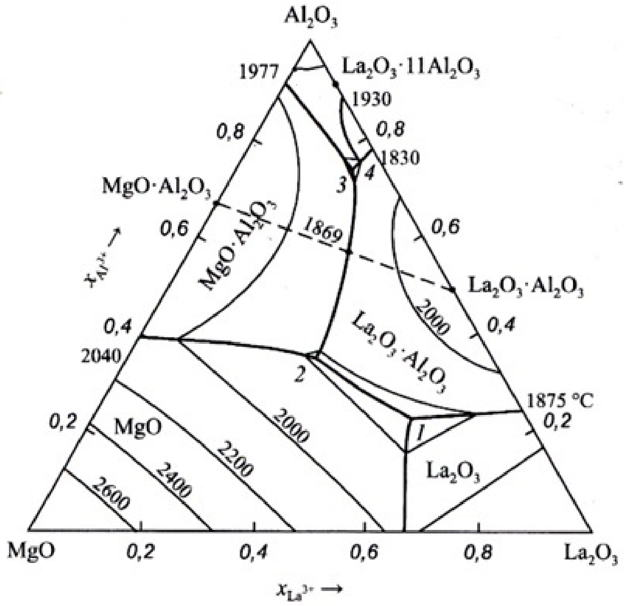


Fig. 1. Phase diagram of La_2O_3-MgO-Al_2O_3 from [14]

rule. The resulting data was also presented graphically. The most reliable data were obtained by measuring the heat capacity of $LaMgAl_{11}O_{19}$ and $SmMgAl_{11}O_{19}$ by differential scanning calorimetry in the high temperature range [17, 18, 19, 20]. These data were presented as the Maier-Kelley equation $C_p(T) = A + B \times T - C/T^2$.

The values of entropy and the changes in enthalpy can be calculated from the known ratios of the heat capacity data:

$$S°(T - 298.15) = \int_{298.15}^{T} \frac{C_p}{T} dT \quad (1)$$

and

$$H°(T) - H°(298.15) = \int_{298.15}^{T} C_p dT. \quad (2)$$

2. Evaluation of the Gibbs energy

To evaluate the thermodynamic stability of hexaaluminates $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm) in the high temperature region, it is necessary to calculate the Gibbs energy of possible reactions for oxides, for which there is data on enthalpies of formation at 298.15 K and on changes in enthalpy and entropy in the high temperature region:

$REMgAl_{11}O_{19} = 0.5\ RE_2O_3 + MgO + 5.5\ Al_2O_3$ (I),

$REMgAl_{11}O_{19} = 0.5\ RE_2O_3 + MgAl_2O_4 + 4.5\ Al_2O_3$ (II),

$REMgAl_{11}O_{19} = REAlO_3 + MgO + 5\ Al_2O_3$ (III),

$REMgAl_{11}O_{19} = REAlO_3 + MgAl_2O_4 + 4\ Al_2O_3$ (IV).

We chose the decomposition reactions into simple oxides, aluminum-magnesium spinel, and $REAlO_3$ aluminates with a perovskite structure because they are present in the triple phase diagram given in [14]. Evaluation by reaction:

$REMgAl_{11}O_{19} = REAl_{11}O_{18} + MgO$ (V)

was impossible due to insufficient data for $REAl_{11}O_{18}$.

For the four above listed reactions, the temperature dependences of the Gibbs energy, which were calculated as the difference between the values for the reaction products and the starting substances, can be presented as follows:

Reaction (I):

$$\Delta_{r(I)}G°(T) = [0.5\Delta_f G°(RE_2O_3, T) + \Delta_f G°(MgO, T) + 5.5\Delta_f G°(Al_2O_3, T)] - \Delta_f G°(REMgAl_{11}O_{19}, T). \quad (3)$$

Reaction (II):

$$\Delta_{r(II)}G°(T) = [0.5\Delta_f G°(RE_2O_3, T) + \Delta_f G°(MgAl_2O_4, T) + 4.5\Delta_f G°(Al_2O_3, T)] - \Delta_f G°(REMgAl_{11}O_{19}, T). \quad (4)$$

Reaction (III):

$$\Delta_{r(III)}G°(T) = [\Delta_f G°(REAlO_3, T) + \Delta_f G°(MgO, T) + 5 \times \Delta_f G°(Al_2O_3, T)] - \Delta_f G°(REMgAl_{11}O_{19}, T). \quad (5)$$

Reaction (IV):

$$\Delta_{r(IV)}G°(T) = [\Delta_f G°(REAlO_3, T) + \Delta_f G°(MgAl_2O_4, T) + 4\Delta_f G°(Al_2O_3, T)] - \Delta_f G°(REMgAl_{11}O_{19}, T). \quad (6)$$

The Gibbs energy of reactions (I-IV) can be expressed as the sum of two components: enthalpy and entropy.

Reaction (I):

$$\Delta_{r(I)}G°(T) = \{[0.5\Delta_f H°(RE_2O_3, T) + \Delta_f H°(MgO, T) + 5.5\Delta_f H°(Al_2O_3, T)] - \Delta_f H°(REMgAl_{11}O_{19}, T)\} - T\{[0.5S°(RE_2O_3, T) + S°(MgO, T) + 5.5S°(Al_2O_3, T)] - S°(REMgAl_{11}O_{19}, T)\}. \quad (7)$$

Reaction (II):

$$\Delta_{r(II)}G°(T) = \{[0.5\Delta_f H°(RE_2O_3, T) + \Delta_f H°(MgAl_2O_4, T) + 4.5\Delta_f H°(Al_2O_3, T)] - \Delta_f H°(REMgAl_{11}O_{19}, T)\} - T\{[0.5S°(RE_2O_3, T) + S°(MgAl_2O_4, T) + 4.5S°(Al_2O_3, T)] - S°(REMgAl_{11}O_{19}, T)\}. \quad (8)$$

Reaction (III):

$$\Delta_{r(III)}G°(T) = \{[\Delta_f H°(REAlO_3, T) + \Delta_f H°(MgO, T) + 5\Delta_f H°(Al_2O_3, T)] - \Delta_f H°(REMgAl_{11}O_{19}, T)\} - T\{[S°(REAlO_3, T) + S°(MgO, T) + 5S°(Al_2O_3, T)] - S°(REMgAl_{11}O_{19}, T)\}. \quad (9)$$

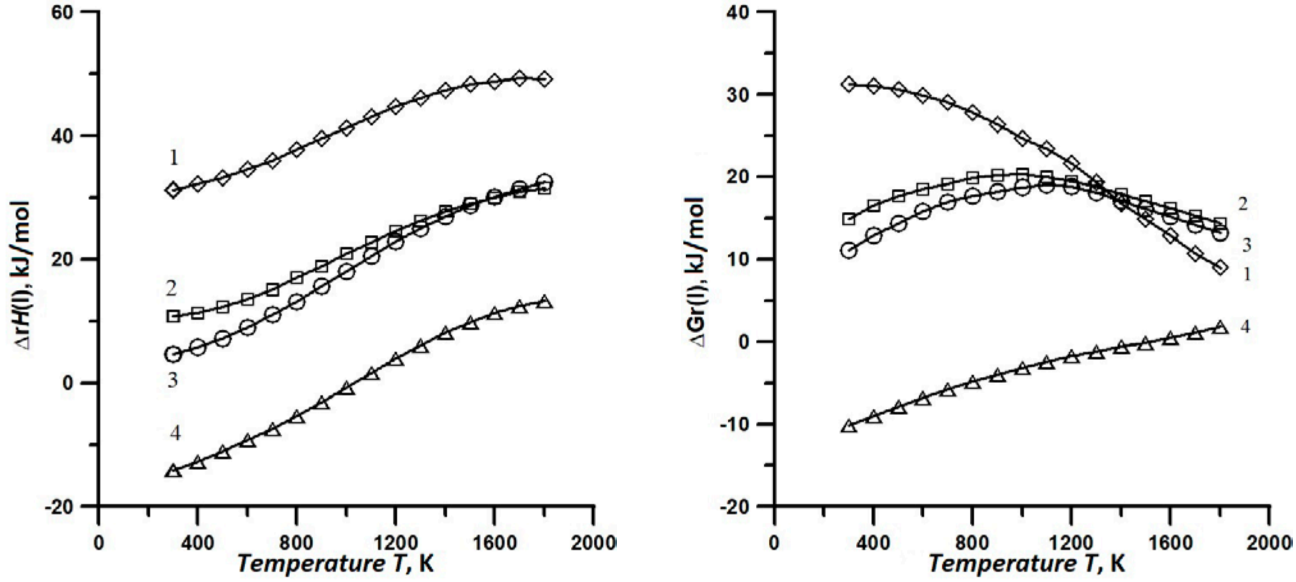
Reaction (IV):

$$\Delta_{r(IV)}G°(T) = \{[\Delta_f H°(REAlO_3, T) + \Delta_f H°(MgAl_2O_4, T) + 4\Delta_f H°(Al_2O_3, T)] - \Delta_f H°(REMgAl_{11}O_{19}, T)\} - T\{[S°(REAlO_3, T) + S°(MgAl_2O_4, T) + 4S°(Al_2O_3, T)] - S°(REMgAl_{11}O_{19}, T)\}. \quad (10)$$

To calculate the enthalpy component over a wide range of temperatures, we needed data on the enthalpies of the corresponding reactions at 298.15 K and the temperature dependences of the changes in enthalpy and entropy for each participant in the reaction.

The thermodynamic values necessary for the calculation were taken from the original articles [17–24] and reference books [25–27]. We found values of enthalpy of formation for $LaAlO_3$ and $PrAlO_3$ perovskites in [28], however, we failed to find data on the temperature dependence of heat capacity. Therefore, calculations were only made for neodymium and samarium compounds. We obtained the estimated enthalpies of formation of hexaaluminates $REMgAl_{11}O_{19}$ (RE = La, Pr, Nd, Sm) with a magnetoplumbite structure by drop calorimetry [29]. The results of calculations of enthalpies and Gibbs energies of type (I–IV) reactions in the temperature range of 298.15–1,800 K are shown in Fig. 2–5.

From Fig. 2, it follows that the values of the Gibbs energy of a type (I) reaction for the lanthanum, praseodymium, and neodymium compounds had positive values in the studied temperature range, while in the case of the samarium compound the sign changed to negative, which may indicate its thermodynamic instability in the region below 1,400 K. However, it should be noted that taking into account the error of determination (about ± 10 kJ/mol), this value can shift to the region of lower temperatures (up to 800 K). There was a general downward trend in thermodynamic stability from lanthanum to samarium.



a b

Fig. 2. Temperature dependences of enthalpy (a) and Gibbs energy (b) of reaction (I) for: *1* – $LaMgAl_{11}O_{19}$, *2* – $PrMgAl_{11}O_{19}$, *3* – $NdMgAl_{11}O_{19}$, *4* – $SmMgAl_{11}O_{19}$

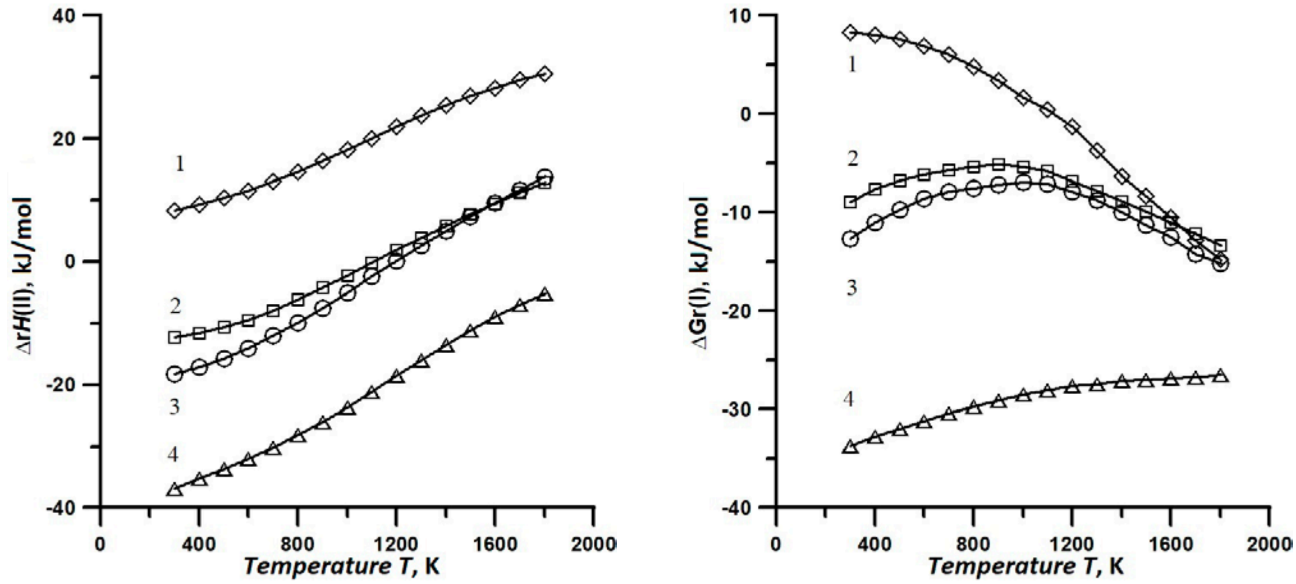


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Fig. 3. Temperature dependences of enthalpy (a) and Gibbs energy (b) of reaction (I) for: *1* – $LaMgAl_{11}O_{19}$, *2* – $PrMgAl_{11}O_{19}$, *3* – $NdMgAl_{11}O_{19}$, *4* – $SmMgAl_{11}O_{19}$

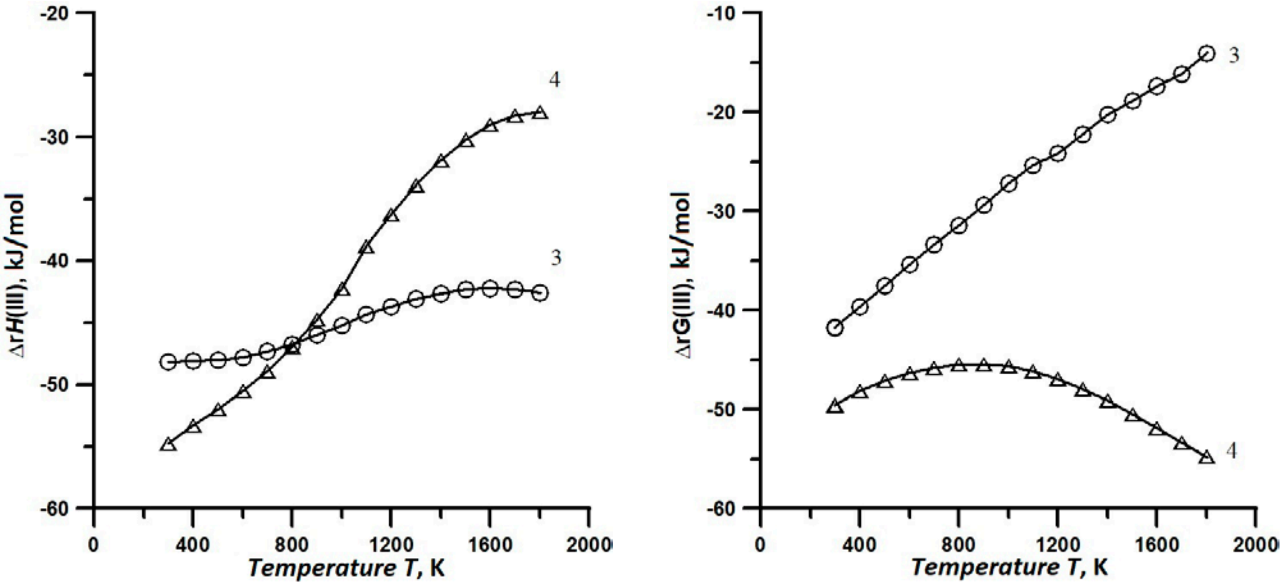

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Fig. 4. Temperature dependences of enthalpy (a) and Gibbs energy (b) of reaction (III) for: *3* – $NdMgAl_{11}O_{19}$, *4* – $SmMgAl_{11}O_{19}$

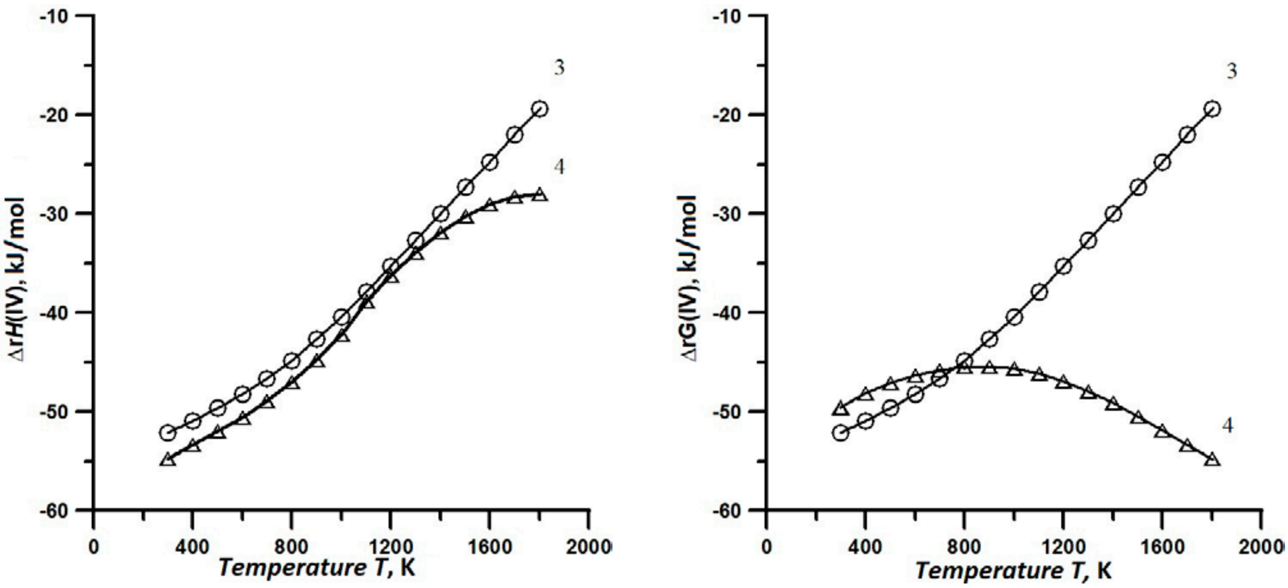

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Fig. 5. Temperature dependences of enthalpy (a) and Gibbs energy (b) (IV) for: *3* – $NdMgAl_{11}O_{19}$, *4* – $SmMgAl_{11}O_{19}$

The values of the Gibbs energy of a type (II) reaction for $LaMgAl_{11}O_{19}$ became negative when the temperature exceeded 1,100 K, which indicates the probability of the reaction. Judging by the temperature dependences of the Gibbs energy for $PrMgAl_{11}O_{19}$, $NdMgAl_{11}O_{19}$, and $SmMgAl_{11}O_{19}$ shown in Fig. 3 and their negative values, a type (II) reaction for these compounds is possible over the entire range of high temperatures.

Judging by the sign of the Gibbs energy of the reaction which involved a decomposition into magnesium and aluminum oxides and $REAlO_3$ perovskites (RE = Nd, Sm), this process is very probable.

Very negative values of the Gibbs energy indicated that a type (IV) reaction for obtaining magnesium-neodymium and magnesium-samarium hexaaluminates from perovskites, spinel, and aluminum oxide should not occur. A significant difference in the type of the Gibbs energy and enthalpy dependencies of type (III) and (IV) reactions can be explained by the influence of the entropy factor.

3. Conclusions

Analysis of the thermodynamic stability of RE magnesium hexaaluminates $REMgAl_{11}O_{19}$ based on the calculation of the Gibbs energy of the decomposition reactions into simple oxides, aluminum-magnesium spinel, and $REAlO_3$ perovskites allowed determining the probability of these reactions over a wide range of temperatures. It was shown that there is influence of enthalpy and entropy factors on the type of temperature dependence of the Gibbs energy of decomposition reactions of hexaaluminates into simpler oxides.

Contribution of the authors

The authors contributed equally to this article.

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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Received 18.07.2024; approved after reviewing 30.07.2024; accepted for publication 16.09.2024; published online 25.12.2024.

Translated by Irina Charychanskaya