ELSEVIER

Journal of Nuclear Materials 247 (1997) 1–6

journal of nuclear materials

Section 1. Vaporization

The high-temperature vaporization/decomposition of actinide materials: fundamental and technological aspects [1]

R.G. Haire *

Chemical and Analytical Sciences Division, Oak Ridge National Laboratory, PO Box 2008, MS-6375, Oak Ridge, TN 37831-6375, USA

Abstract

The unique and variable chemistry exhibited across the actinide series offers a scientifically challenging group of elements. Investigations of their high-temperature materials science and thermodynamics can provide important insights into fundamental actinide science and generate critical information for technological applications. The wide variations in the physicochemical properties and the availability of the various actinide elements necessitates special experimental techniques be employed, even when investigating a single property (e.g., the enthalpy of vaporization). Experimental approaches range can from classical to novel methods to permit studies of the rarer and the more intensely radioactive members. We have developed special equipment and techniques to investigate the high-temperature, physicochemical properties of several actinides through Md, as well as the precursor elements, radium and actinium. These experimental approaches employed both alloys and solid solutions of compounds, in addition to the pure elements themselves. The vaporization/decomposition of compounds and the sublimation of the elements, together with the associated thermodynamics, are the main issues. Results for several of the actinide elements, some of the techniques employed and systematic comparisons of the thermochemical aspects of bonding are discussed to give a fuller understanding of the series as a whole. © 1997 Elsevier Science B.V.

---

1. Introduction

The physicochemical properties of the actinides, including the high-temperature behaviors of the elements and their compounds, vary considerably across the series. These variations result to a large extent from significant changes that occur in the role of the 5f-electrons. In this regard, plutonium and einsteinium appear to occupy pivotal positions in the series. At normal pressures and temperatures, the 5f electrons are no longer considered itinerant after plutonium, while einsteinium becomes the first divalent actinide metal. This divalency arises because of the greater energy required for promoting 5f-electrons to a d-bonding state. Thus, electronic structure controls the chemistry and much of the physics of the actinide elements and their compounds. In the discussion at hand, the fundamental issues are: (1) understanding how experimental parameters, such as temperature, alter the unperturbed electronic states; and/or (2) how bonding affects high-temperature behaviors.

In general, the radioactive nature and toxicity of the actinides present experimental obstacles but additional difficulties, due to the scarcity and more intense radioactivity, are encountered in the second half of the series. Although larger quantities of transplutonium elements have become available in recent years, the quantities of these elements still limit significantly the experimental studies that can be performed with them and special techniques are often required. For the heaviest of the actinide members, their extreme scarcity often prevents direct measurement (e.g., Knudsen effusion techniques) of high-temperature properties, even with miniaturized or modified techniques. Then an indirect (e.g., thermochromatography) techniques may become the viable technique.

We have conducted studies of the vaporization of transplutonium elements through mendelevium, and the

---

* Tel.: +1-423 574 5007; fax: +1-423 574 4987; e-mail: rghaire@ornl.gov.

[1] Research sponsored by the Division of Chemical Science Office of Basic Energy Sciences, US Dept. of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Research Corporation.

PII S0022-3115(97)00024-X

vaporization/decomposition of their oxides through Fm, via Knudsen effusion techniques. The primary goal and the motivation for performing such high-temperature studies with the actinides are to promote an understanding of the basic science of the actinides, as well as enhance the technological knowledge of these elements, through systematic comparisons. A fundamental goal is to pursue an understanding of the entire series in terms of electronic configurations. Technological goals concentrate on materials science issues, especially for the elements, U through Cm.

This paper describes the experimental considerations, approaches and the results obtained to date for several actinide materials. The results obtained, together with previously existing data, allow systematic comparisons of thermochemical behaviors to be formulated in terms of electronic configurations and bonding.

2. Experimental

2.1. Materials

The transuranium elements employed in these studies were obtained either directly or indirectly (e.g., as daughter products from radioactive decay) from processing campaigns at the High Flux Isotope Reactor at Oak Ridge National Laboratory. The metals were obtained by techniques described elsewhere [1,2]. Alloys were prepared either by melting the components together or by codistilling and condensing multiple metals. Oxides samples, both as a single oxide and as solid solutions of oxides, were prepared from chloride solutions of the elements by oxalate precipitations; and then calcining the oxalate salts to obtain an oxide product. Initially these calcining conditions were 1275 K in air but subsequent treatments involved vacuum or hydrogen and temperatures from 1175–1875 K in order to adjust the oxide's stoichiometry.

2.2. Instrumental

X-ray powder diffraction analysis (molybdenum K_α, radiation) was the main tool used to assess the metal and oxide products in the studies. Interplanar distances of the products were refined and cell parameters determined via standard computer least squares programs.

The vaporization behaviors of both the metallic and oxide materials where determined using one of three different Knudsen effusion systems designed for handling different quantities of radioactive materials. The main system was of more conventional design and employed 1–10 mg samples. It consisted of a target collection mechanism, an in-line alpha pulse analyzer and a 1000 AMU mass spectrometer. The maximum temperature attainable was 3300 K. This system was used for studying both the elements and their compounds; it was the primary systems for oxides since it had the capability for mass analysis and identification.

The two other effusion instruments were designed for maximum collection geometry; each was used for specific quantities of material. The smaller of the two was for samples consisting of only thousands of atoms; the other was for multi-microgram quantities. Neither incorporated a mass spectrometer, since only tracer levels of materials were studied with them. These systems relied on radioactive counting analyses for identification and for determining the isotope and its mass. The main difference between these systems was the geometry, the target devices used and the maximum temperatures that were attainable (1500–1800 K). Their collection geometries were 3–5 × larger than that of the first system described. A schematic of the intermediate system is published [3], and it is also appropriate for the smallest system. The general Knudsen cell operation, target collection procedures and calculation of pressure from the experimental data are adequately discussed in Ref. [4].

An useful approach for determining the vapor pressure of one element during the simultaneous vaporization studies of two materials (e.g., two actinides), where the vaporization behavior of one component has been established, is given below. The approach employs the standard equation for calculating vapor pressures from vaporization data but makes use of the fact that many of the experimental parameters may cancel out, when two similar materials are being vaporized simultaneously. For this technique to be valid, each material should effuse independently of the other and condense on the targets with the same coefficient of condensation. If the atomic weights are close (e.g., within few percent, as with some of the actinides), then the pressure of one material can be calculated from a ratio of the masses condensed on the targets, and the known pressure of the other. Thus, for an alloy or solid solution of A and B, where the temperature dependence of B is known

$$P_{\mathrm{A,mixture}} = [\text{mass A}/\text{mass B}][P_{\mathrm{B,mixture}}].$$

The mass of each isotope on the targets is attained by radiochemical counting techniques. This technique has been useful for the parent–daughter systems (e.g., Fm–Cf; Es–Bk, etc.), where the parent is relatively short lived.

3. Results and discussion

3.1. High-temperature vaporization: elements and alloys

One of the important bulk properties of an element is its enthalpy of vaporization, which can be a measure of its cohesive energy. The number of bonding electrons (metallic valence) can be inferred from the magnitude of this enthalpy, which allows systematic comparisons to be made between elements. For example, trends in the vaporization enthalpies of the 3d-, 4d- and 5d-transition metals show a

Table 1
Enthalpy of vaporization for selected elements

Element	Enthalpy, kJ mol^{-1} at 298 K	Ref.	Assigned metallic valence
Eu	176	[3]	2
Yb	155	[3]	2
Es	133	[5]	2
Fm	143	[4]	2
Ba	192	[6]	2
La	422	[6]	3
Gd	418	[6]	3
Lu	405	[6]	3

definite correlation with their d-electron occupation number. With the actinides, the trend in vaporization enthalpies is more variable than with the transition metals, due to the significant and important role of promotion energies and the occurrence of hybrid bands involving f-electrons, as found with the lighter actinides. Nevertheless, comparisons of actinide enthalpies are still useful. The variation found with the divalent actinide metals is much smaller, as it is mainly the s-electrons that are bonding; f-electron promotion energies are then not contributing factors. Thus, a more uniform and similar enthalpy of vaporization (133 to 175 kJ mol^{-1}; Eu [5], Yb [5], Es [4] and Fm [3]) is observed with these divalent metals, as seen in Table 1. The higher actinides, Md and No and Ba [6] (divalent precursor of the lanthanide series) and Ra, would also be expected to have similar enthalpies based on these concepts. The enthalpies of vaporization for several of these elements and the trivalent La, Gd and Lu metals, are given in Table 1. Extrapolated values for Md and Md can be seen in Fig. 1. There is interest in Ra (precursor of the actinides) to determine experimentally if its enthalpy is indeed comparable to these other divalent metals. A very difficult but important experiment will be to determine the enthalpy of vaporization of Ac, whose enthalpy would be expected to be similar to that of La [6] (nominally, 400 kJ mol^{-1}). Both Ra and Ac would be best studied via radiochemical techniques, such as we have used for the transcalifornium elements. This approach would avoid, or at least minimize, complications that would be encountered from the strong radiation emulating from these elements and/or their daughter products. The large variation in enthalpies observed for the trivalent actinide metals, a variation of over 200 kJ/mol, results from changes in the f-electron promotion energies when going from element to element.

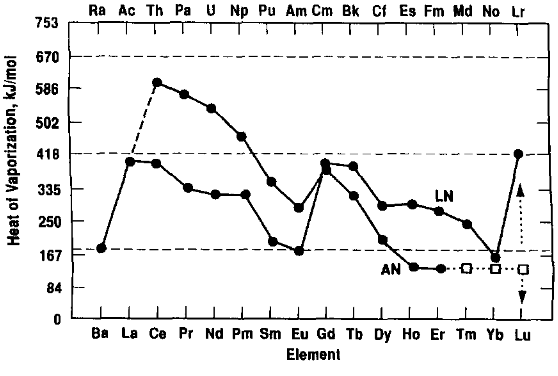

Fig. 1. Variation of the enthalpy of vaporization with element for the 4f- and 5f-series of elements.

A common technique for determining the enthalpy of vaporization for a metal is to determine the equilibrium partial pressure of its atomic vapor as a function of temperature (e.g., the Knudsen effusion method), where the intensity of an atomic beam effusing from a cell is determined as a function of temperature. Many actinide metals have been studied by this approach [7]; we have performed measurements on pure actinide metals and their compounds through Cf using this approach and employing milligram or less quantities of them. With the next three elements, Es, Fm and Md, availability decreases to microgram to nanogram scale and then to only a thousand-or-so atoms. For the study of these elements, we have developed a technique that employ alloys. With even smaller quantities (from hundreds down to only a few atoms) it is necessary to adopt another experimental approach, such as the thermochromatograpy technique. This technique will be discussed briefly in a later section.

We have developed different instrumental systems to study actinide metals and compounds via the Knudsen effusion technique for these different quantities of material (see the experimental section). Smaller quantities of actinide material are best studied using dilute actinide alloys. For these smaller quantities, it is important to maximize the collection geometry to conserve the material and avoid its depletion during the study. It is important to emphasize that in studies involving alloys, the actinide pressures are measure over an alloy and it is necessary that its vaporization from the alloy be related in a known way to that from the pure actinide. This situation is best approximated when ideal alloys are formed, and care must be taken in choosing the actinide's host material to achieve this condition. For the divalent metal Es, Yb is a good host; for Am, trivalent La would be a suitable host. However, this selection is not always straight forward. With the better defined lanthanide system, where activities of the alloy's components have been measured, it has been found that even with the relatively similar trivalent metals, Sm and Y, or Sm and Gd, neither couple forms an ideal alloy. It appears in these cases that Sm metal has sufficient divalent character to avoid forming ideal alloys with Y and Gd [8].

The alloy technique has been used by us to determine the enthalpies of vaporization of Es [4] and Fm [3]. In the case of Es, Yb was selected as the host; with Fm, both Yb and Sm hosts were employed. In the case of Fm, the Sm alloys gave slightly lower calculated pressures over the alloy than were found over the Yb alloy, although the enthalpies determined by both second and third law treatments were identical for both systems within experimental limits [3]. Whether the lower pressures obtained with the Sm host indicated some effect on the metallic valence of Fm (e.g., induced some partial trivalent character to the divalent metal) could not be ascertained. The enthalpies for Es and Fm are shown in Fig. 1 and listed in Table 1.

The three dashed horizontal dividing lines shown in Fig. 1 represent zones that can be assigned as representing apparent metallic valences of two, three or greater than three for the f-metals. The very large enthalpy range for the trivalent zone comes about from the effect of f-electron promotion energies on the enthalpies of vaporization. Elements falling above this trivalent zone (e.g., > 420 kJ/mol), have f-electrons participating in bonding (e.g., hybrid band formation), suggesting the involvement of more than three bonding electrons). The plot of the enthalpies of vaporization for the elements in Fig. 1 provides some indication of the elements' apparent metallic valences.

Recently, we have attempted to extend our determinations to the next higher actinide, Md, and determine experimentally its enthalpy of vaporization [9] using nominally a thousand atoms of it. Although these initial experiments were themselves unsuccessful, due mainly to the limited amount of material and the its lower specific activity (Md-257 isotope), we have shown with other actinides that this experiment is feasible for this mass level. The enthalpy indicated in Fig. 1 for Md is therefore only an estimate; the method for making this estimate will be discussed in a later section.

Experimental attempts have not been made for No or Lr. Given both the availability and half-lives for isotopes of these elements, it is not feasible to attempt direct vaporization measurements, as described here, or probably even the indirect thermochromatography technique to be discussed in a latter section. There is little question that No should be a divalent metal and it is very likely that Lr will behave as a trivalent metal, even considering the possibility for a monovalent state for it brought about by relativistic effects. Approximate values for No and Lr suggested in Fig. 1 are also based on extrapolations.

3.2. *Thermochromatography measurements*

We have not performed thermochromatography experiments on the actinide elements but this important technique has relevance to some of our findings and extrapolations and is therefore discussed briefly.

The thermochromatographic and the vaporization techniques both provide information about the electronic states of actinide elements; however, the former is capable of working with smaller amounts of material. The main feature of the thermochromatographic method is that it involves adsorption enthalpies rather than vaporization enthalpies. Since these techniques can overlap in scope, and the enthalpies of adsorption can be obtained with smaller quantities of material, it is highly desirable to be able to inter-compare values obtained by both vaporization and adsorption. Previous reports of studies involving enthalpies of adsorption [10,11] provided values for Cf, Es, Fm and Md. An earlier approximate correlation by us [12] between the enthalpies of vaporization and adsorption of Cf, Es and Fm, allowed an extrapolation (of 134 to 142 kJ mol^{-1}) for the enthalpy of vaporization for Md. This is the Md value plotted in Fig. 1.

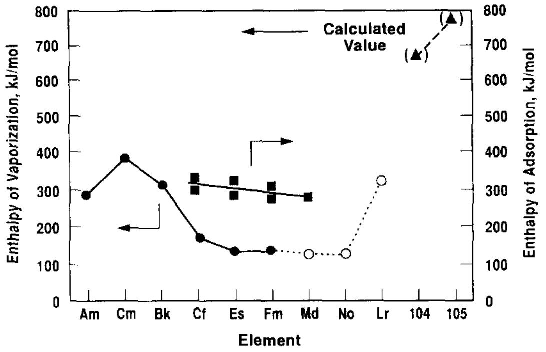

Fig. 2. Comparison of the enthalpies of vaporization and adsorption for selected transplutonium elements.

Recently, additional studies on the enthalpy of adsorption of actinides have been reported and tabular comparisons made between the enthalpies of vaporization and adsorption [13,14]. Calculated adsorption values based on theoretical considerations have been given for selected actinides [14]. A comparison of the enthalpies of vaporization and adsorption that have been reported for Cf, Es and Fm, together with the calculated enthalpies of vaporization for elements 104 [15] and 105 [16] is shown in Fig. 2. For comparison, the enthalpy of vaporization of Zr, (element 104 has been assigned as a group V element, falling under Hf in the periodic table) is 584 kJ mol^{-1}, similar to the enthalpy reported for element 104. The enthalpy for element 105 is expected to be even larger [16] (see Fig. 2).

One drawback to the thermochromatography technique for estimating the metallic valence is that an element may yield different adsorption enthalpies when different surfaces are used for collection. This makes the assignments of metallic valence based on adsorption (e.g., which value should be used in assigning a metallic valence?) and the correlation of enthalpies of vaporization and adsorption enthalpies, more difficult This variation is apparent in the case of Cf metal, where the largest deviation in the enthalpy of adsorption was noted for the different collection surfaces [10,11]; Cf could be assigned as a divalent or a trivalent metal depending on the collection material used. This variation with Cf metal is in accord with the magnitude of its promotion energy (f^{n}s^{2} to f^{n-1}s^{2} — divalent to trivalent metal). As the promotion energy for Cf is very close to the divalent–trivalent threshold, Cf should be the most susceptible of these three elements, Cf, Es and Fm, to show a variation with the collection surface. Studies of 'bulk' quantities of Cf metal have shown definitively that this element is a trivalent metal [17]. However, this variation in adsorption enthalpies establishes that interesting differences in electronic behavior can be observed for these different divalent metals on the atomic scale.

Another potential difficulty in such thermochromatographic studies is assuring that the few volatile atoms generated are metal, and remain as metal, during transport and adsorption. That is, they must not collide with oxygen or other residual materials in the gas phase, or upon adsorption, to form monoxides or other compounds, which would then generate enthalpies for materials other than the elemental atoms. Given the number of atoms involved, even with great care, it would be difficult to remove totally such residuals, and one is then faced in part with collision probabilities.

An interesting point in this regard is the potential stability of the monoxides of the divalent actinide metals, Es, Fm, Md and No, in the gas phase. Our recent studies of oxides systems, discussed in a subsequent section, have shown that the monoxides of Cf, Es, Fm and Md (and very likely No) are unstable with respect to atomic vapor at elevated temperatures. This situation makes the existence and maintenance of atomic vapor over monoxide more probable in such adsorption experiments. However, this situation changes for Lr and presumable for elements 104 and 105, where once formed their monoxides should be the more stable species in the vapor. That is, their monoxide dissociation energies should be much larger. Formation of oxides or other compounds during condensation on the collection surfaces (e.g., reaction with surface atoms of the collector or foreign atoms residing on the collector's surface) is a different consideration.

3.3. Dependence of oxide vaporization on electronic structure

Relating the effect of electronic configurations and f-electron promotion energies of the actinide elements provide insights not only into the high-temperature behaviors of the elements but also into those of their compounds. It is the stability of particular electronic structures which determine oxidation states and the stoichiometries that are exhibited for the oxides (or other compounds). Less obvious is the fact that the f-electron promotion energies for the elements play a major role in the vaporization/decomposition of actinide oxides at elevated temperatures, especially in the case of the monoxides. The latter is based on concept that it is necessary to have a ds-orbital for monoxide bonding: that is, a d-orbital must be available (e.g., ds^2 rather than a fs^2 configuration) on the metal atom for formation of the monoxide [18]. Formation of this d-orbital at the expense of an f-electron involves the f-electron's promotion energy, and the monoxide bond is therefore 'weakened' by an equivalent of this promotion energy. Thus, the larger the actinide f-electron's promotion energy the smaller the monoxide's dissociation energy, and the more probable the formation of atomic rather than monoxide vapor during the vaporizing of actinide oxides. The major factors for determining the primary products during vaporization of the oxides is the magnitude of the element's monoxide and dioxide dissociation energies. Establishing this relationship between monoxide dissociation energy and the f-electron promotion energy for the Am through Lr monoxides has provided both predictive and correlative methodologies for the vaporization of these oxides [19].

Our recent efforts have been focused on comparing systematically the vaporization and/or decomposition behaviors of the higher actinide oxides. Considerable work, much of it published, had been done on the lighter actinide oxides; much less has been done on the transplutonium oxides.

The variation in actinides' binary oxide stoichiometries is much greater than that observed with the lanthanides. In examining the actinide oxides, it can be observed that after Np in the series, the highest binary actinide oxide solid formed is the dioxide. This stoichiometry is known for the actinides from Th through Cf, but at temperatures above 1500 K, where vaporization is normally encountered, essentially only the dioxides of Th, U, Np and Pu are sufficiently stable to be considered as the solid phase from which vaporization proceeds. Therefore, only the sesquioxides of Am through Fm are expected as the primary phase from which vaporization occurs.

At the risk of oversimplification, the lighter actinide oxides tend to vaporize to produce mixtures of atomic vapor, monoxides and dioxides (mainly UO_2 and ThO_2), while the transplutonium oxides produce monoxides, atomic vapors or mixtures of these. The magnitude of the monoxide's dissociation energy determines the ratio of monoxide to atomic vapor observed in the gas phase over the solid. Table 2 lists established and calculated [19] monoxide dissociation energies for the actinides. When monoxide dissociation energies are above 600 kJ mol^{-1}, the more probable it becomes for monoxide vapor products, while with lower dissociation energies, the products are expected to be atomic vapor. Mixtures of atomic and monoxide vapor are expected when the dissociation energy is in the range of 600 kJ mol^{-1}.

One of the primary goals of such vaporization studies is to derive enthalpies of vaporization and formation, and the free energies of formation of the oxides. Space does not permit such discussion of the individual actinide oxides' thermodynamics. However, the enthalpies of formation of the transplutonium sesquioxides have been found to be similar to one another, having enthalpies that range from -1600 to -1700 kJ mol^{-1}.

3.4. Technological aspects: vaporization of actinide compounds

Fundamental information on the actinides provides important input for technological avenues, and is critical for understanding numerous applications involving these ele-

Table 2
Dissociation energies for the monoxides[a]

Oxide	D_0 (kJ mol^{-1})	Oxide	D_0 (kJ mol^{-1})
LaO	794	AcO	(794)
CeO	786	ThO	891
PrO	736	PaO	788
NdO	699	UO	774
PmO	(711)	NpO	736
SmO	569	PuO	703
EuO	469	AmO	(550)
GdO	711	CmO	728
TbO	690	BkO	(598)
DyO	611	CfO	(498)
HoO	602	EsO	(460)
ErO	602	FmO	(443)
TmO	510	MdO	(418)
YbO	397	NoO	(268)
LuO	665	LrO	(665)

[a] Ref. [19]. ( ) = calculated values.

ments. Given the quantities of individual actinides on hand and current technological efforts, the major interest is concentrated on the actinides from U through Cm. A considerable amount of information on vaporization behavior has been published on the U and Pu systems, due to the military and commercial interests in these elements. Less has been published about Am compounds.

There are many examples of vaporization processes that are important to technology. A few examples of the interests are: the preferential vaporization of Pu in the PuC system to form Pu_2C_3; the separation of Am and Pu from a fluoride matrix via preferential vaporization of PuF_6 [20]; the preferential loss of Am from (Am, Pu)N [21]; and the vaporization of Am from oxide matrices.

Expanding on the latter subject, the applicable reactions for the vaporization of pure Am oxide are

$$2AmO_2 \rightarrow Am_2O_3 + \tfrac{1}{2}O_2 \quad \text{and} \quad Am_2O_3 \rightarrow 2Am + 3/2O_2.$$

At temperatures where vaporization can be considered, there is an appreciable reduction of the dioxide to the hexagonal sesquioxide, so that this material is the primary solid phase from which vaporization is expected. Given that the dissociation energy of AmO is nominally 550 kJ mol^{-1} [19], the vaporization product will be atomic Am vapor rather than AmO_2 or AmO. In principle, one collects Am metal. The volatilization of the oxide can begin at relatively low temperatures (e.g., 1400 K), which can be encountered in certain technological applications and must be addressed.

4. Concluding remarks

The high-temperature vaporization behaviors of the elements and compounds have and will continue to provide important insights to fundamental science, as well as providing valuable information for technological issues. It should be clear from the information provided here that the changing role of the f-electrons (including bonding and nonbonding roles) and the f-electron promotion energies are critical factors in the physical and thermodynamic properties of the actinides and their compounds.

When classical experimental approaches are not feasible (e.g., due to the quantities of material), novel techniques can often be found to permit the desired science to be pursued. In some the work described here, it was shown that enthalpies of vaporization for the elements and their oxides provide important thermodynamic information applicable to both fundamental and technological science, and this information can be obtained directly or indirectly for the majority of the actinides.

References

[1] R.G. Haire, in: Actinides in Perspective, ed. N. Edelstein (Pergamon, New York, 1982) p. 309.
[2] R.G. Haire, in: Actinides-85, ed. M. Beauvy (Elsevier, Lausanne, 1986) p. 379; and references therein.
[3] N.E. Tropp, in: The Chemistry of the Rare Earth Elements (Elsevier, New York, 1965) p. 134.
[4] R.G. Haire, J.K. Gibson, J. Chem. Phys. 91 (1989) 7085.
[5] P.D. Kleinschmidt, J.W. Ward, G.M. Matlock, R.G. Haire, J. Chem. Phys. 81 (1984) 473.
[6] O. Kubaschewski, C.B. Alcock, in: Metallurgical Thermochemistry (Pergamon, New York, 1979) p. 358.
[7] J.W. Ward, P.D. Kleienschmidt, R.G. Haire, D. Brown, in: Lanthanide and Actinide Chemistry and Spectroscopy, ACS Symposium Series 131, ed. N. Edelstein (American Chemical Society, Washington, DC, 1980) p. 199.
[8] C.E. Lundin, A.S. Yamamoto, J.F. Nachman, Acta Metall. 13 (1965) 149.
[9] R.G. Haire, M. Gensini, J.K. Gibson, R. Lougheed, unpublished results.
[10] S. Hubener, I. Zvara, Radiochim. Acta 31 (1982) 89.
[11] S. Hubener, I. Zvara, Radiochem. Radioanal. Lett. 44 (1980) 79.
[12] R.G. Haire, J.K. Gibson, J. Radioanal. Nucl. Chem. 143 (1990) 35.
[13] S. Hubener, B. Eichler, M. Schadel, W. Bruchle, K.E. Gegorich, D.C. Hoffman, J. Alloys Compounds 213&214 (1994) 429.
[14] S. Taut, S. Hubener, B. Eichler, H.W. Gaggeler, M. Schadel, E. Jager, W. Bruchle, in: 1995 Annual Report, Forschungszentrum Rossendorf, Institute of Radiochemistry, p. 101.
[15] E. Johnson, B. Fricke, J. Phys. Chem. 95 (1991) 7082.
[16] V. Pershina, B. Fricke, G.V. Ionva, E. Johnson, J. Phys. Chem. 98 (1994) 1482.
[17] J.W. Ward, P.D. Kleinschmidt, R.G. Haire, J. Phys. (Paris) C4 (1979) 233.
[18] E. Murad, D.L. Hildenbrand, J. Chem. Phys. 73 (1980) 4005.
[19] R.G. Haire, J. Alloys Compounds 213&214 (1994) 185.
[20] T.R. Mills, L.E. Reese, J. Alloys Compounds 213&214 (1994) 360.
[21] T. Ogawa, T. Ohmichi, A. Maeda, Y. Arai, Y. Suzuki, J. Alloys Compounds 224 (1995) 55.