Electrochemical studies on plutonium in molten salts

G. Bourgès [a],[*], D. Lambertin [a], S. Rochefort [a], S. Delpech [b], G. Picard [b]

[a] *CEA—Centre d'études de Valduc, 21 120 Is sur Tille, France*
[b] *Laboratoire d'Electrochimie et de Chimie Analytique (UMR7575, CNRS), ENSCP, 11 rue Pierre et Marie Curie, 75231 Paris, France*

Abstract

Electrochemical studies on plutonium have been supporting the development of pyrochemical processes involving plutonium at CEA. The electrochemical properties of plutonium have been studied in molten salts – ternary eutectic mixture $NaCl–KCl–BaCl_2$, equimolar mixture NaCl–KCl and pure $CaCl_2$ – and in liquid gallium at 1073 K. The formal, or apparent, standard potential of Pu(III)/Pu redox couple in eutectic mixture of $NaCl–KCl–BaCl_2$ at 1073 K determined by potentiometry is equal to -2.56 V (versus Cl_2, 1 atm/Cl^- reference electrode). In NaCl–KCl eutectic mixture and in pure $CaCl_2$ the formal standard potentials deduced from cyclic voltammetry are respectively -2.54 V and -2.51 V. These potentials led to the calculation of the activity coefficients of Pu(III) in the molten salts. Chronoamperometry on plutonium in liquid gallium using molten chlorides – $CaCl_2$ and equimolar NaCl/KCl – led to the determination of the activity coefficient of Pu in liquid Ga, $\log \gamma = -7.3$. This new data is a key parameter to assess the thermodynamic feasibility of a process using gallium as solvent metal. By comparing gallium with other solvent metals – cadmium, bismuth, aluminum – gallium appears to be, with aluminum, more favorable for the selectivity of the separation at 1073 K of plutonium from cerium. In fact, compared with a solid tungsten electrode, none of these solvent liquid metals is a real asset for the selectivity of the separation. The role of a solvent liquid metal is mainly to trap the elements.
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1. Introduction

Pyrochemical separations, involving molten salt and metal media, by liquid/liquid extraction or electrorefining, are studied for nuclear defense and civil applications [1–3]. These processes concern actinide separations, however, lanthanides, such as cerium, are often used as surrogates. The first steps of a pyrochemical process development consist in studying the chemistry of the elements to be separated in the solvent media. Activity coefficients of the solutes in the two phases, that described the solvent–solute interactions, are important thermochemical parameters to predict separation efficiency and to assess the solvents influence.

Actinides/lanthanides separation in fluoride molten salt and metallic Al–Cu system is considered for spent fuel pyroprocessing by CEA [4] in the frame of international research programs.

CEA has been operating pyrochemical processes in chloride media for more than 40 years for plutonium metal preparation and recovery. The determination of the thermochemical parameters of plutonium in molten chlorides and metal media by electrochemical measurements has been supporting the development of these processes. In the early studies, Pu(III)/Pu electrochemical system was investigated in the ternary $NaCl–KCl–BaCl_2$ [5] eutectic mixture by potentiometry. Later, Pu(III)/Pu system was studied in equimolar NaCl–KCl eutectic mixture and in pure $CaCl_2$ [6] by cyclic voltammetry. As solvent metal such as gallium could have potentialities for the recovery of plutonium from metallic scraps, the activity coefficient of plutonium dissolved in gallium was determined [7] using cyclic voltammetry and chronoamperometry.

2. Experimental

For all experiments, the feed chlorides, $CaCl_2$, $BaCl_2$, NaCl and KCl (ACS reagents) were heated at $200\,^\circ$C under vacuum and then were melted and argon sparged to remove water. Pure commercial gallium and plutonium metal pieces prepared at CEA/Valduc were used as metal feeds. The molten gallium–plutonium metal phase was obtained by melting pieces of pure pluto-

* Corresponding author. Tel.: +33 3 8023 4397; fax: +33 3 8023 5219.
E-mail address: gilles.bourges@cea.fr (G. Bourgès).

nium and gallium covered by the molten salt at 1123 K. A stirring sequence at 1073 K provided a perfect dissolution of the plutonium before the measurements. About 500 g of gallium were used in these experiments. Pu(III) was introduced in the molten salt by anodic oxidation of liquid Pu metal at a controlled potential and the resulting concentration was determined by coulometry. The concentration of Pu(III) was checked by gamma spectrometry on the solid salt after the experiments.

The experiments were carried out in a glove box under inert atmosphere. The chlorides, ternary mixture of $NaCl36/KCl36/BaCl_2 28$ (molar fraction), equimolar NaCl–KCl mixture or $CaCl_2$ and the metal were melted in a magnesia crucible placed inside a vertical stainless steel furnace under dry argon at atmospheric pressure. A classical three electrodes cell was used for the electrochemical investigations and the measurements were performed with a slight stirring of the melt. The reference electrode was made of a silver wire (1 mm diameter) dipped into a porous alumina or a quartz tube containing a solution of silver chloride in the considered chloride (from 0.02 to 0.08 molar fraction of AgCl) [4,8]. Two 1 mm diameter tungsten wires were used as working and counter electrodes. For liquid metal working electrodes (gallium or plutonium), the metal was introduced at the bottom of the crucible and the contact was made with a tungsten wire isolated from the molten salt using a magnesia or alumina tube. The electrochemical measurements were performed using an AUTOLAB PGSTAT30 potentiostat coupled with a PC computer for NaCl–KCl and $CaCl_2$ studies and a TACUSSEL PRT 200 potentiostat for NaCl–KCl–$BaCl_2$ studies.

3. Results and discussion

3.1. Electrochemical studies of plutonium in eutectics NaCl–KCl–BaCl₂, NaCl–KCl and in pure CaCl₂

Cyclic voltammograms were recorded at a tungsten electrode in equimolar NaCl–KCl and $CaCl_2$ at 1073 K (Figs. 1 and 2) and in NaCl–KCl–$BaCl_2$ at 1000 K (Fig. 3) containing or not Pu(III) ions.

In NaCl–KCl melt containing Pu(III), Fig. 1 shows two cathodic peaks Ic and IIc at potentials respectively equal to -2.8 V and -2.9 V (versus Cl_2, 1 atm/Cl^-). Many studies have demonstrated the plutonium (III) reduction into plutonium metal in one-step in chloride melts [3,9,10]. So, the cathodic peaks Ic and IIc are attributed respectively to the reduction of Pu(III) into Pu and the reduction of Na^+ into Na. The cathodic limit of equimolar NaCl–KCl solvent salt was previously determined
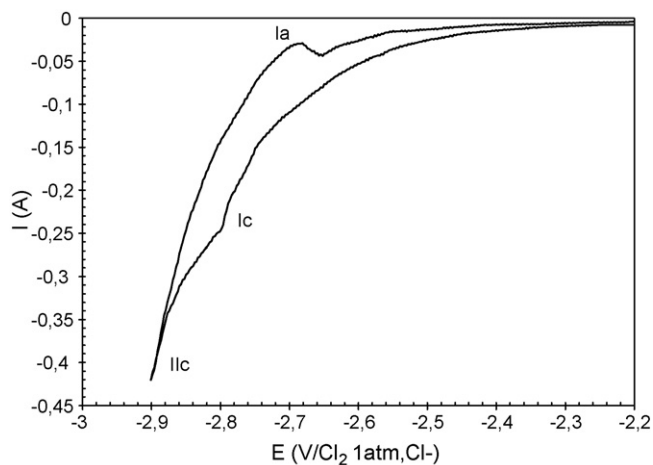

Fig. 2. Cyclic voltammogram for plutonium trichloride in $CaCl_2$ at 1073 K, scan rate 0.1 V/s, $xPuCl_3 = 1.1 \times 10^{-3}$, surface area = 0.31 cm².

and corresponds to $E_{Na(I)/Na} = -3.28$ V (versus Cl_2, 1 atm/Cl^-) [7,8]. The reduction potential of Na^+ (peak IIc) is more positive with Pu(III). This potential increase may be due to the presence of plutonium metal on the electrode before the Na^+ reduction leading to the formation of a Pu–Na compound (activity of Na is not equal to 1). Another hypothesis is the presence of a complex anion in the melt, e.g. $PuCl_6^{3-}$ that may change the acidity of the melt and probably the activity of NaCl at the interface. This potential increase results in plutonium sodium co-reduction at low Pu(III) activity. McCurry and Moy [11] reported a similar phenomenon.

Voltammograms of Pu(III) in $CaCl_2$ at 1073 K (Fig. 2) and in NaCl–KCl–$BaCl_2$ at 1000 K (Fig. 3) exhibit well-defined peaks corresponding to Pu(III)/Pu redox system. In these media, the difference between the plutonium trichloride reduction and solvent salt reduction (reduction of Na^+ or Ca^{2+}) is larger than in NaCl–KCl.

The formal (or apparent) standard potential of the Pu(III)/Pu system in the melts can be deduced by using the following

Fig. 1. Cyclic voltammogram for plutonium trichloride in equimolar NaCl–KCl at 1073 K, scan rate 0.1 V/s, $S = 0.31$ cm², $xPuCl_3 = 1.05 \times 10^{-3}$ and for pure equimolar NaCl–KCl (background, $S = 0.287$ cm²).

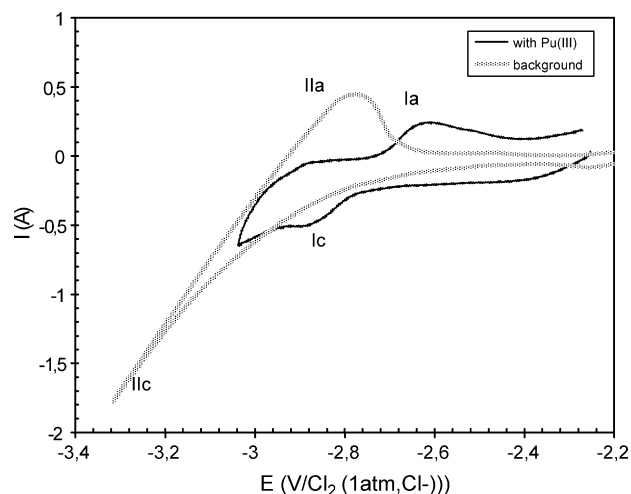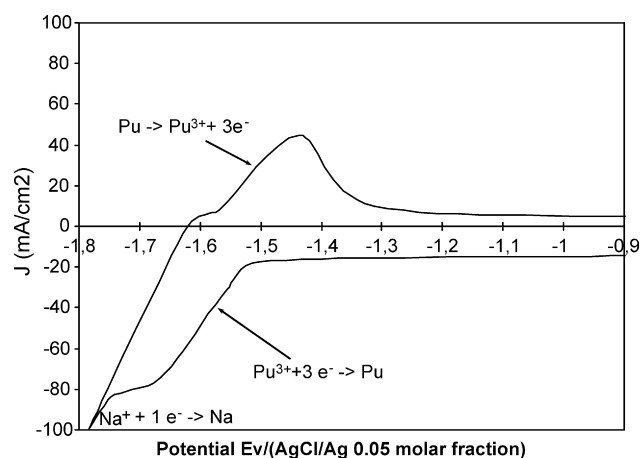Fig. 3. Cyclic voltammogram for Pu trichloride in NaCl/KCl/$BaCl_2$ at 1000 K, scan rate 0.3 V/s, $S = 1$ cm², $xPuCl_3 = 5 \times 10^{-3}$.

expression derived from the Nernst relation:

$$E_{eq} = E^*_{Pu(III)Pu} + \frac{2.3RT}{3F} \log x_{Pu(III)} \qquad (1)$$

where E_{eq} is the measured equilibrium potential, $E^*_{Pu(III)/Pu}$ the formal standard potential of the Pu(III)/Pu in the considered salt, $x_{Pu(III)}$ the mole fraction of Pu(III) in the molten salt and $R = 8.31$ J/mol K and $F = 96{,}485$ C/mol.

The obtained value of $E^*_{Pu(III)/Pu}$ allows to calculate the activity coefficient of Pu(III) in the molten salt, $\gamma_{Pu(III)}$, using the following relation where the Gibbs free energy of formation $\Delta G^\circ_{PuCl_3}$ of the supercooled liquid is determined from the literature [12]:

$$\log(\gamma_{Pu(III)}) = \frac{3F E^*_{Pu(III)/Pu} - \Delta G^\circ_{PuCl_3}}{2.3RT} \qquad (2)$$

The formal standard potential of Pu(III)/Pu redox system in NaCl–KCl–BaCl$_2$ have been determined by potential measurements at various Pu(III) concentrations and temperatures ranging between 973 and 1123 K of the following galvanic cell [5]:

$$Pu|Pu(III)_{NaCl-KCl-BaCl_2}||Ag(I)_{NaCl-KCl-BaCl_2}|Ag$$
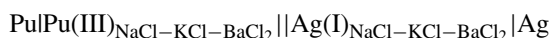
In these measurements, the working electrode consists of liquid plutonium in contact with the molten salt that contains plutonium ions. Pu(III) additions in the molten salts were obtained by anodic oxidation sequences. The experimental isotherms (Fig. 4) of the potential versus logarithm of the Pu(III) concentration fit onto straight lines with slopes close to the value of $2.3RT/3F$ in relation (1) at the corresponding temperature. This observation confirms that Pu(III) ions are in equilibrium with Pu metal and that the activity coefficient of Pu(III) in the melt tends to be constant at low concentrations. The expression of the formal standard potential of Pu(III)/Pu redox system as a function of the temperature deduced from these measurements is:

$$E^*_{Pu(III)/Pu} = -3.34 + 7.28 \times 10^{-4}T \quad (\text{versus } Cl_2, \ 1\,atm/Cl^-)$$
$$(3)$$

For equimolar NaCl–KCl and CaCl$_2$ at 1073 K, the values of the Pu(III)/Pu redox formal potentials have been estimated from cyclic voltammograms. The equilibrium potential, E_{eq}, was approximated from the zero current intercepts of the reverse
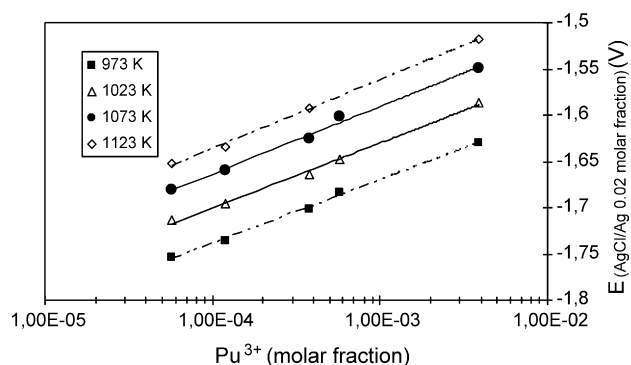

Fig. 4. Potentiometry of Pu(III)/Pu couple in NaCl/KCl/BaCl$_2$.

Table 1
Values of formal potential of Pu(III)/Pu in various molten chloride mixtures at 1073 K (V vs. Cl$_2$ (1 atm)/Cl$^-$, mole fraction scale) and their corresponding activity coefficients

Molten chlorides	$E^*_{Pu(III)/Pu}$		$\log(\gamma_{Pu(III)})$	
	Skiba & Silin	This work	Skiba & Silin	This work
KCl	−2.605		−1.42	
NaCl–KCl–BaCl$_2$		−2.56		−0.79
NaCl–KCl	−2.585	−2.54 ± 0.01	−1.14	−0.507
NaCl	−2.513		−0.126	
CaCl$_2$		−2.51 ± 0.01		−0.084

scans of the cyclic voltammetry [6,13] and led to the determination of the formal potential $E^*_{Pu(III)/Pu}$ value by using the expression (1).

The formal standard potentials of Pu(III)/Pu redox couple and the corresponding calculated activity coefficients at 1073 K in eutectics NaCl–KCl–BaCl$_2$, NaCl–KCl and in pure CaCl$_2$ are summarized in Table 1. The uncertainty in E values obtained by the apparatus used is about 1 mV, therefore the uncertainties on experimental E_{eq} are estimated at 10 mV. Our results are almost consistent with the results reported by Skiba and Silin for media based on NaCl and KCl [10,14].

The activity coefficient is characteristic of the complexation of plutonium ions in the melt at a given temperature. Complexation of Pu(III) ions appears to be much more important in KCl based media than in pure CaCl$_2$ where Pu(III) ions can be considered as not solvated.

A perfect drying of the salt is not possible in industrial conditions. Consequently, the resulting oxides ions concentration in molten salt must be considered. The potential—p(O^{2-}) Pourbaix-type diagrams were built from previous results and from literature data in order to give a comprehensive view of the properties of plutonium in such media [15]. This approach showed that Pu(III) solubility is higher in CaCl$_2$ than in equimolar NaCl–KCl [6].

3.2. Thermochemical properties of plutonium in liquid gallium at 1073 K

Cyclic voltammograms, recorded in molten CaCl$_2$ on a liquid gallium working electrode containing or not plutonium, are reported in Fig. 5. Curve (a) without plutonium, that represents the electrochemical window on gallium metal in CaCl$_2$ media, exhibits two peaks Ia and IIc that are attributed, respectively, to the oxidation of gallium metal into gallium chloride (mono or trichloride) and to the reduction of Ca^{2+} into calcium metal in liquid gallium. In presence of plutonium (curve (b)), the peaks I$'$a and I$'$c are attributed, respectively, to the oxidation of Pu (dissolved in liquid gallium) into Pu(III) and to the reduction of Pu(III) into Pu(Ga). The peak II$'$c probably results from the formation of intermetallic Pu–Ga solid compounds on the electrode because this peak is independent of Pu(III) concentration. The deposition a such intermetallic solid compounds have been reported by Finne et al. for the reduction of Pu(III) on a Ga
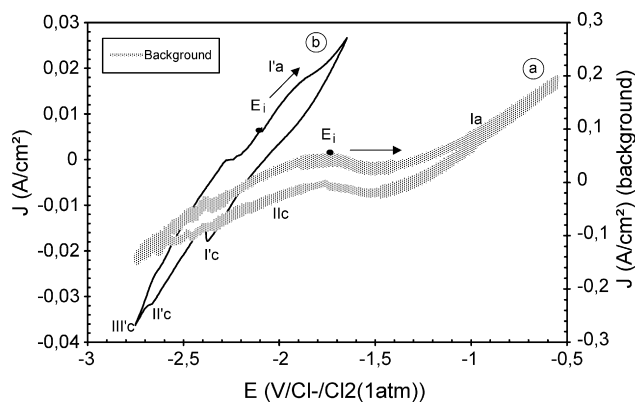
Fig. 5. Cyclic voltammetry on liquid gallium in CaCl₂ at 1073 K, curve (a): xPu(Ga) = 0, xPuCl₃ = 0, curve (b): xPu(Ga) = 0.0129, xPuCl₃ = 0.0011.

Fig. 6. Intensity–potential curve in CaCl₂ at 1073 K obtained from chronoamperograms, xPu(Ga) = 0.0129, xPuCl₃ = 0.0011.

electrode at 833 K [16]. In these experiments, no plutonium was dissolved in the gallium.

These redox systems seem to be irreversible so we cannot deduce any characteristic potential value from cyclic voltammograms. In order to obtain equilibrium potential values, chronoamperograms have been performed and are reported as $I = f(E)$ curves at various time with CaCl₂ (Fig. 6) and with the equimolar NaCl–KCl (Fig. 7) molten salts. The curves present an anodic wave before the anodic limit IIa. These redox systems are, respectively, attributed to the oxidation of Pu (dissolved in the gallium) into Pu(III) and to the oxidation of Ga into Ga(I) or Ga(III). The equilibrium potential at zero current can be obtained from current–potential curves given in Figs. 6 and 7. This potential is set by the redox system Pu(III)/Pu$_\mathrm{Ga}$ given by the following expression:

$$E_{\mathrm{Pu(III)/Pu_{Ga}}} = E^*_{\mathrm{Pu(III)/Pu}} + \frac{2.3RT}{3F} \log \frac{x\mathrm{Pu(III)}}{x\mathrm{Pu_{Ga}}}$$
$$- \frac{2.3RT}{3F} \log \gamma\mathrm{Pu_{Ga}} \qquad (4)$$

where $E^*_{\mathrm{Pu(III)/Pu}}$ is the formal potential of Pu(III)/Pu in the chloride media ($E^*_{\mathrm{Pu(III)/Pu}} = -2.51$ V in CaCl₂ and $E^*_{\mathrm{Pu(III)/Pu}} = -2.54$ V in NaCl–KCl), $\gamma\mathrm{Pu_{(Ga)}}$ is the activity coefficient of plutonium and $x\mathrm{Pu_{Ga}}$ the molar fraction of plutonium in liquid gallium.

The activity coefficients of plutonium in liquid gallium calculated by using relation (4) are reported in Table 2. The values obtained with two molten chloride salts are of the same order of magnitude. This is consistent with that the activity coefficient in the metal phase does not depend on the nature of the solvent salt. Consequently, we can retain a value of log(γPu$_\mathrm{Ga}$) equal to -7.3 ± 0.5 at 1073 K for low concentrations of plutonium in liquid gallium (near 0.01 molar fraction). For an exhaustive study of the potentialities of gallium as solvent metal for pyroprocesses, this data could be completed by values of the activity coefficient of plutonium in gallium near saturation by using the protocol proposed by Finne et al. [16,17]. In this work, values of log(γPu$_\mathrm{Ga}$) at saturation between 733 and 833 K are deduced from measurements on liquid Ga in equilibrium with a solid PuGa intermetallic compound.

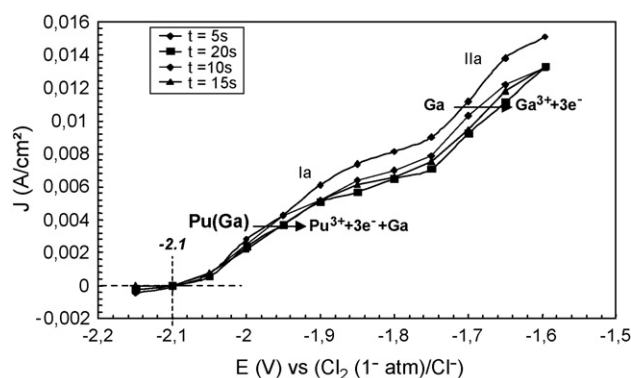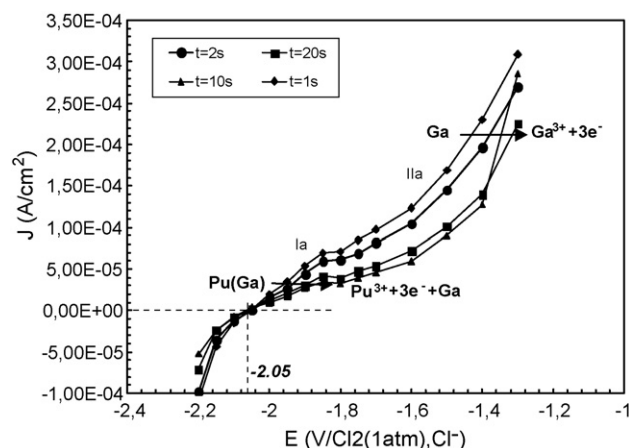Fig. 7. Intensity–potential curve in NaCl/KCl at 1073 K obtained from chronoamperograms, xPu(Ga) = 0.0137, xPuCl₃ = 6.65 × 10⁻⁴.

Table 2
Values of molar fraction ($x_\mathrm{Pu(Ga)}$) of metal plutonium in liquid gallium and its corresponding log(γPu$_\mathrm{Ga}$) at 1073 K

Molten chlorides	xPu(Ga)	log(γPu$_\mathrm{Ga}$)
CaCl₂	0.0129 ± 0.0005	−7.5 ± 0.3
NaCl–KCl	0.0137 ± 0.0005	−7.2 ± 0.3

3.3. Influence of the solvent metal: application to plutonium cerium separation

The thermochemical properties of cerium in gallium were considered previously to plutonium in the frame of this work [6]. The value of the logarithm of the activity coefficient of Ce in liquid Ga at 1073 K has been determined to log(γCe$_\mathrm{Ga}$) = −8.27 ± 0.15, witch is consistent with Lebedev's result: (log(γCe$_\mathrm{Ga}$) = −7.9) [18]. As activity coefficient of Pu in liquid Ga was not available in literature, gallium has been compared to other liquid metals such as aluminum, bismuth and cadmium for the separation of an actinide, i.e. plutonium from a lanthanide, i.e. cerium.

To point out the influence of metal solvent on thermodynamic selectivity, we have calculated the difference of potential (i.e. electrochemical selectivity) between plutonium and cerium, by
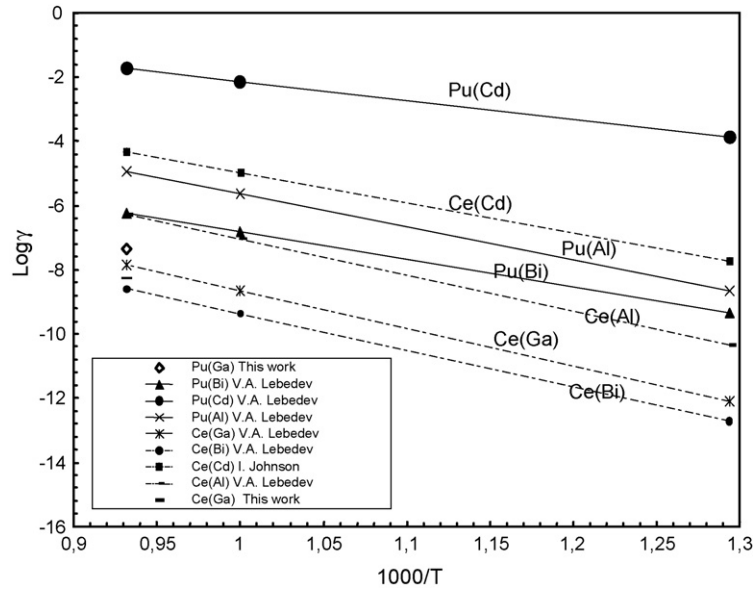
Fig. 8. Temperature dependence of activity coefficients of plutonium in bismuth, cadmium, aluminium, gallium (this work) and activity coefficients of cerium in gallium, bismuth, aluminium, cadmium.

using our present results on liquid gallium and the published values for aluminum, bismuth and cadmium [18,19]. The activity coefficients of Pu and Ce in these liquid metals are reported in Fig. 8.

This difference of potential (electrochemical selectivity) between plutonium and cerium with a metallic solvent (MS) can be expressed as:

$$E_{Pu} - E_{Ce} = \Delta E_{Pu-Ce}^* + \frac{2.3RT}{3F} \log \left(\frac{x Pu(III)_{salt} x Ce_{(MS)}}{x Ce(III)_{salt} x Pu_{(MS)}} \right)$$
$$+ \frac{2.3RT}{3F} \log \left(\frac{\gamma Ce_{(MS)}}{\gamma Pu_{(MS)}} \right) \tag{5}$$

where ΔE_{Pu-Ce}^* is the difference of the formal standard potentials of Pu(III)/Pu an Ce(III)/Ce in the molten salt, $\Delta E_{Pu-Ce}^* = +0.47\,V$ in equimolar NaCl–KCl [20,7], and $\gamma Pu_{(MS)}$ and $\gamma Ce_{(MS)}$ are the activity coefficients of plutonium and cerium in metal solvent.

The selectivity of the separation between plutonium and cerium is:

$$S(Pu/Ce) = \left(\frac{x Ce(III)_{salt} x Pu_{(MS)}}{x Pu(III)_{salt} x Ce_{(MS)}} \right) \tag{6}$$

At equilibrium, the selectivity between plutonium and cerium becomes:

$$\frac{2.3RT}{3F} \log S(Pu/Ce) = \Delta E_{Pu-Ce}^* + \frac{2.3RT}{3F} \log \left(\frac{\gamma Ce_{(MS)}}{\gamma Pu_{(MS)}} \right) \tag{7}$$

The influence of the solvent metal on the selectivity between plutonium and cerium in a molten chloride is described by the following parameter:

$$\Delta E_{(Pu-Ce)MS} = \frac{2.3RT}{3F} \log \left(\frac{\gamma Ce_{(MS)}}{\gamma Pu_{(MS)}} \right) \tag{8}$$

The values of $\Delta E_{(Pu-Ce)MS}$ as function of temperature obtained from published values and from our results (Fig. 8) are reported in Fig. 9. If $\Delta E_{(Pu-Ce)MS}$ is high in absolute value, the selectivity decreases. At 1073 K, the use of liquid gallium is better than bismuth and cadmium. It is more difficult to conclude about the comparison of gallium and aluminum because the gap between their $\Delta E_{(Pu-Ce)MS}$ is small and the uncertainties should be taken into account. At lower temperatures (near 873 K), for witch the plutonium and cerium concentrations approach or exceed the solubility limits in the solvent metal, the selectivity between plutonium and cerium decreases in cadmium and is nearly constant in bismuth. For aluminum, which solidify at 933 K, the selectivity increases.
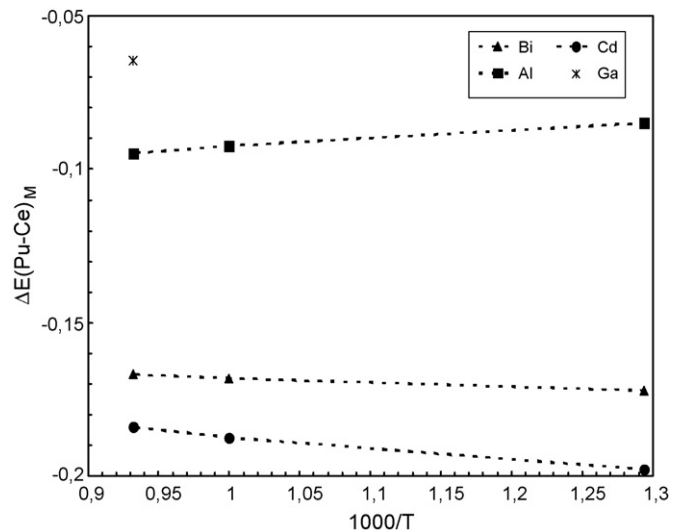

Fig. 9. Electrochemical selectivity between plutonium and cerium in gallium, cadmium, aluminium and bismuth as a function of temperature.

For an inert solid electrode such as tungsten, the activity coefficients of Pu and Ce are equal to 1, and the selectivity depends on the difference between the formal potentials in the molten salt. As all values of $\Delta E_{(Pu–Ce)MS}$ are negative, we can deduce that, by considering thermodynamic data, a solid electrode as tungsten is more favorable for the separation of Pu from Ce than all the considered liquid metal electrodes.

4. Conclusion

In the frame of developments of pyrochemical processes involving plutonium, the electrochemical properties of plutonium have been studied in the ternary eutectic mixture NaCl–KCl–BaCl$_2$, the equimolar mixture NaCl–KCl and pure CaCl$_2$ at 1073 K. The formal standard potential of Pu(III)/Pu redox couple in eutectic mixture of NaCl–KCl–BaCl$_2$ determined during earlier studies by potentiometry is equal to -2.56 V. In equimolar NaCl–KCl and in pure CaCl$_2$ the standard potential deduced from cyclic voltammetry are respectively -2.54 V and -2.51 V.

Electrochemical investigations on plutonium diluted in liquid gallium using molten chlorides showed that gallium can be used as solvent metal for pyrochemical processes involving plutonium at 1073 K. In particular, a selective extraction of plutonium from gallium by anodic oxidation is thermodynamically possible. The deposition of plutonium on a liquid gallium cathode is also possible with CaCl$_2$ as molten salt with a careful control of cathode potential.

Activity coefficients of cerium and plutonium in gallium have been deduced from these electrochemical studies. Comparison with other solvent liquid metals such as cadmium, bismuth, aluminum suggests that gallium, with aluminum, is more favorable for a Pu/Ce separation at 1073 K. In fact, none of these solvent metals is a real asset for the selectivity of such separation on the thermochemical point of view. The role of solvent metal is mainly to trap the elements.

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