Study by XRD of the lattice swelling of PuGa alloys induced by self-irradiation

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Abstract

Plutonium aging leads to the creation of decay products, such as americium, uranium and helium and self-irradiation defects such as vacancies, vacancy clusters, self-interstitials and helium bubbles. As these self-irradiation defects accumulate in plutonium–gallium alloys, the lattice parameters of the material change. Thus, this work is an X-ray diffraction (XRD) study of the lattice parameter changes, such as kinetics and amplitude, as a function of self-irradiation dose for non-homogenized and homogenized samples. The results have shown no incubation time before the beginning of lattice swelling. Moreover, whereas the lattice swelling amplitude seems to be influenced by the gallium segregation to the border of grain in non-homogenized samples, it is not strongly influenced by the gallium concentration. An average lattice parameter increase of about 4.5×10^{-3} Å is observed for homogenized alloys at saturation. This equilibrium is achieved after a dose of 0.1 dpa. A discussion of possible causes leading to these observations and their effects is presented.
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1. Introduction

Plutonium metal has six different phases between room temperature and its melting point (640 °C) at ambient pressure. However, the high-temperature δ-phase (face-centered cubic structure), stable from 315 to 457 °C, can be held at room temperature by alloying plutonium with a few atomic percent of a so-called 'δ-phase stabilizer' element such as gallium, aluminum, cerium, or americium. How-

ever, the plutonium–gallium alloy stabilized in the δ-phase at room temperature is metastable and undergoes an extremely slow eutectoid decomposition to a mixture of α-plutonium and Pu_3Ga. The stabilizing mechanisms induced by such additions are still not understood [1–4].

Additionally, plutonium aging must be taken into account. Indeed, the α-decay leads to the creation and recoil of an energetic helium nucleus (~5 MeV) and a uranium nucleus (~86 keV), both producing defects such as vacancies, interstitials and clusters of these. The range of the α-particle (helium nucleus) in the plutonium crystal is about 10 μm and this induces a displacement cascade

which is 0.8 μm in diameter, whereas the range of the uranium nucleus is much shorter, about 12 nm, and the diameter of the cascade is about 7.5 nm [5]. Combinations between helium atoms and vacancies may also occur to stabilize vacancies or vacancy clusters as well as to form helium bubbles [6].

The type and concentration of stable defects strongly depend on temperature since it governs defect mobility. Therefore, several studies were performed at low temperature to limit the recovery of self-interstitials, vacancies or vacancy clusters and thus to analyze the influence of these defects on plutonium properties. Regarding the swelling measured by X-ray diffraction, the results completely differ depending on the nature of the plutonium phase. Whereas swelling is observed at low temperature for pure plutonium, which is explained by an increase in the amount of defects in the metal [7–10], a contraction of the lattice is observed for plutonium alloys stabilized in the δ-phase [11–13]. In the latter case, the contraction was attributed to an electronic effect. More precisely, it was suggested that a plutonium atom in an interstitial position promoted the 5f–6d hybridization of plutonium atoms leading to a delocalization of the 5f electrons and consequently to a decrease in the atomic volume. To investigate this further, electrical resistivity measurements as a function of temperature were performed on a plutonium–gallium 3.3 at.% alloy. This experiment provided an identification of the different stages of migration for the different defects: stage I occurs from 20 up to 45 K, stage II from 45 to 150 K, stage III from 150 to 180 K, stage IV from 180 to 250 K, and finally stage V from 250 to 310 K [14]. Thus, vacancies migration at temperatures above 150 K produces vacancy clusters, which means that the migration of vacancies and interstitials induced by a displacement cascade is very high at room temperature.

Only a few experimental studies have examined room temperature aging in plutonium alloys stabilized in the δ-phase. X-ray diffraction experiments performed on plutonium–aluminum alloys with aluminum contents of 11.5 and 12.8 at.% showed a relative lattice swelling of 10^{-3} after aging at room temperature for 10 years. This corresponds to an increase of about 4.5×10^{-3} Å in the lattice parameter [15]. Another work performed by Chebotarev and Utkina [16] on plutonium–gallium alloys with different gallium contents found that the lattice swelling reached a maximum value after 2 or 3 years

and that the relative lattice swelling at saturation linearly increased with the gallium content in the alloys. According to Chebotarev and Utkina [16], the lattice swelling is reversible since annealing the aged sample above 150 °C restores the initial lattice parameter. More recent work by Lawson confirmed Chebotarev's finding that the increase in lattice parameter is proportional to the gallium content [17].

The purpose of this work is to precisely study the changes of lattice parameter with time for plutonium–gallium alloys with different gallium contents. This provides a better understanding of the kinetics and amplitude. Experimental details are reported in Section 2, measurements are given in Section 3, and a discussion about the origin of the lattice swelling is detailed in Section 4.

2. Experimental details

2.1. Samples description

As freshly cast samples were not available for plutonium–gallium alloys stabilized in the δ-phase with gallium contents of 3.4 and 6 at.%, two aged alloys were heat treated (called HAPUGA34 and HAPUGA6). Table 1 shows the age in dpa (displacements per atom; 0.1 dpa corresponds to about 1 year). The helium concentration was calculated from the isotopic content of the plutonium, and the americium content was determined by chemical analysis. Samples were machined into cylinder with diameters of 10 mm and heights of 2 mm for XRD analysis. A 200 h heat treatment at 460 °C was then performed on all the samples under high vacuum condition (2×10^{-7} mbar) to nearly fully homogenize the solute distribution and to restore the crystalline structure. This heat treatment did not affect the helium, uranium, americium and other impurities content that were initially present in the samples.

Table 1
Age, theoretical concentration in helium and americium concentration in samples HAPUGA6 and HAPUGA34 before heat treatment (after 1 year corresponding to 0.1 dpa, 40 appm of helium atoms are generated)

Alloys	Age (dpa)	Theoretical He concentration (appm)	Americium concentration (appm)
HAPUGA6	3.08	1047	6070
HAPUGA34	1.750	592	2720

Additionally, to study the influence of gallium distribution on lattice swelling, two freshly cast plutonium–gallium samples with a gallium content of 1.9 at.% underwent a 10 h heat treatment at 460 °C to *partially* homogenize the gallium distribution and to ensure that both samples were δ-monophased. Because much of the gallium was still segregated through the grains, these samples were referred to as 'cored'. An additional heat treatment of 200 h at 460 °C was performed on one of the cored samples to nearly fully homogenize the gallium distribution (the cored freshly cast sample is called CFPUGA19 and the homogenized freshly cast one HFPUGA19).

To remove surface oxides and other possible impurities before each XRD analysis samples were electropolished at a voltage of 40 V in a cooled bath (90% ethylene glycol and 10% nitric acid) at a temperature of 266 K. Then, samples were stored under vacuum between each XRD analysis. All plutonium handling was performed in a glove box with a circulating nitrogen supply.

2.2. Experimental conditions

XRD experiments were performed using a classical $\theta/2\theta$ diffractometer (Brüker D8 Advance) with copper radiation. Since the diffractometer was not in a glove box, samples had to be confined in a sealed polymer film (called Rilsan®) before characterization in order to avoid contamination. XRD analyses were performed in the reflection mode with an X-ray beam energy of 8.048 keV, which probed a maximum of 2 μm (calculated for 90% of the contribution to the diffracted beam). Thus, only the surface of samples contributed to the XRD diagrams. To improve the counting statistics, the sample was rotated during the data acquisition. The detector used was a positive sensitive detector (Braun), integrating an angular range of 6°, with a 12 μm thick nickel foil used to remove the K_β radiation. In order to limit the beam divergence of the primary radiation beam, a slit with a width of 0.2 mm was used. The diffraction diagrams were recorded at ambient temperature with a step time of 0.3 s and a step size of 0.015° from 30° up to 130° in 2θ angle. Before each XRD series of analyses, the calibration of the device was verified using NIST standard Silicon (Si 640 c). To determine the peak position, the diffraction peaks corresponding to the δ-phase of the plutonium were fitted with a mathematical function called split pseudo-voigt using TOPAS 2.1 [18] soft-

ware. The lattice parameters were then refined using the program Celref 3.0 [19], which is a crystal lattice parameter refinement program for powder XRD that is based on a least squares refinement method. The accuracy of each lattice parameter was determined from the value of the mean square deviation obtained after refinement.

3. Results

3.1. Phase analysis

XRD diagrams recorded for HAPUGA6, HAPUGA34, CFPUGA19 and HFPUGA19 samples before aging are presented in Fig. 1. Whatever the gallium content is, plutonium–gallium alloys are completely stabilized in the δ-phase. As a matter of fact, no additional phase transition has been observed during aging (until about 0.4 dpa). The diffraction peak observed for all sample at 36.5° (2θ) is induced by the presence of the semi-crystalline polymer Rilsan® film using as confinement.

3.2. Initial lattice parameter

The lattice parameters, which are determined on homogenized alloys (HFPUGA19, HAPUGA34 and HAPUGA6), are presented and compared with the previous published data from Ellinger et al. [20] in Fig. 2. Our experimental values are in a good agreement with Ellinger's data and confirm the negative deviation from the Vegard's law. This phenomenon is explained by an electronic effect consisting in a 5f(Pu) and 4p(Ga) hybridization, i.e. 5f electrons states delocalization. More precisely, it occurred by replacing 6d–5f hybridization by 4p–5f hybridization when a plutonium atom is substituted to a gallium one. Moreover, the agreement between Ellinger's data and our results for HAPUGA34 and HAPUGA6 samples highlights the total reversibility of the lattice swelling during heat treatment even if helium, uranium and americium atoms remain in the crystalline lattice after annealing. However, a difference in the initial value of lattice parameter could be noticed between homogenized and cored samples (CFPUGA19 and HFPUGA19) as illustrated in Fig. 2. The lattice parameter for the cored alloy appears higher than for the homogenized one, suggesting that the mean gallium concentration seen by XRD for cored sample seems to be smaller. As determined by EPMA (electron probe micro analysis), the cored samples
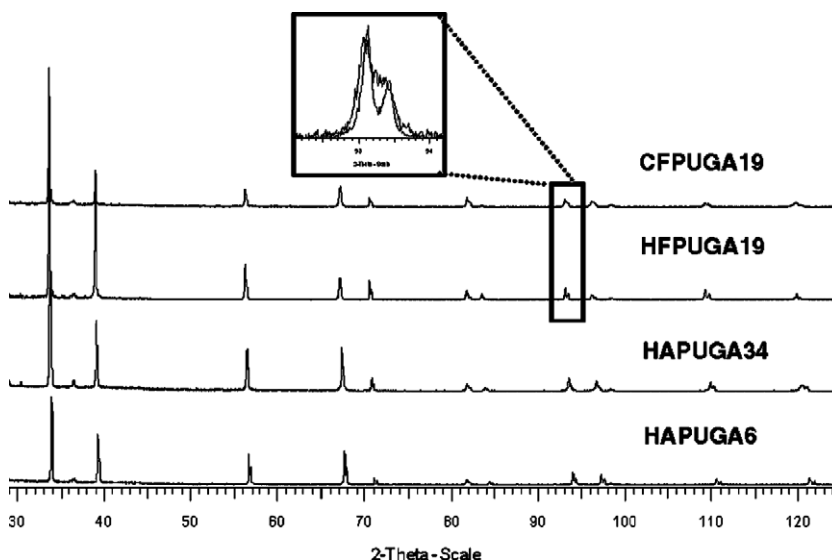
Fig. 1. XRD diagrams of HFPUGA19, CFPUGA19, HAPUGA34 and HAPUGA6 samples before aging.
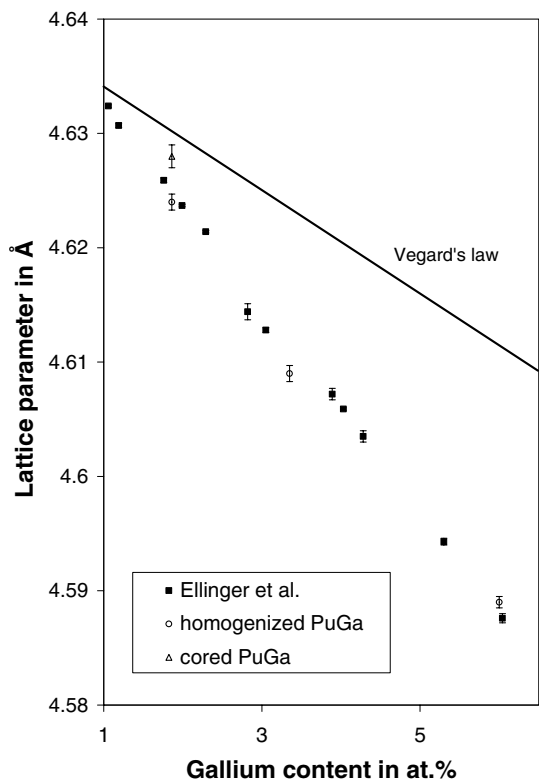

Fig. 2. Lattice parameter of plutonium–gallium alloys as a function of gallium content. Our experimental values (open symbols) are compared to Ellinger's data [20] confirming the negative deviation from the Vegard's law and showing a difference in lattice parameter between cored and homogenized alloys.

show a gradient into the gallium distribution [21,22], the center of the grain being more concentrated than the grain boundaries. Then, for XRD analysis, each diffraction Bragg peak is a convolution of sub peaks resulting from a same lattice plane but with a lattice plane spacing slightly different due to the inhomogeneous distribution of gallium through the grain. Thus, for a given lattice plane, the position and the profile of the corresponding diffraction Bragg peak depends on a few parameters, such as the position, the profile and the intensity of each sub peak, which are influenced by defects and lattice microdistortions. Considering that a same profile is observed for each sub peak, the resulting position of the diffraction Bragg peak depends only on the position and the intensity of these sub peaks. The intensity of sub peaks is related to the diffracting volume corresponding to a given gallium content. Since the gallium content radial distribution decreases from the center to the grain boundary, it results that the diffracting volume corresponding to the low gallium content (at the grain boundary) is enhanced leading to a stronger intensity for the sub peaks corresponding to the lattice plane of the gallium depleted zones. Thus, the convolution of those sub peaks gives resulting diffraction peaks broader than these corresponding to homogenized alloys with an asymmetric shape, which mean position tends towards the low 2θ angles and consequently towards greater lattice

parameters. This effect is characterized by a broadening as well as a shift of the Bragg peaks to the lower angles as shown in Fig. 1. So, the 'smaller apparent' gallium content analyzed by XRD in the cored alloys can be simply interpreted as an experimental artifact, which has to be considered
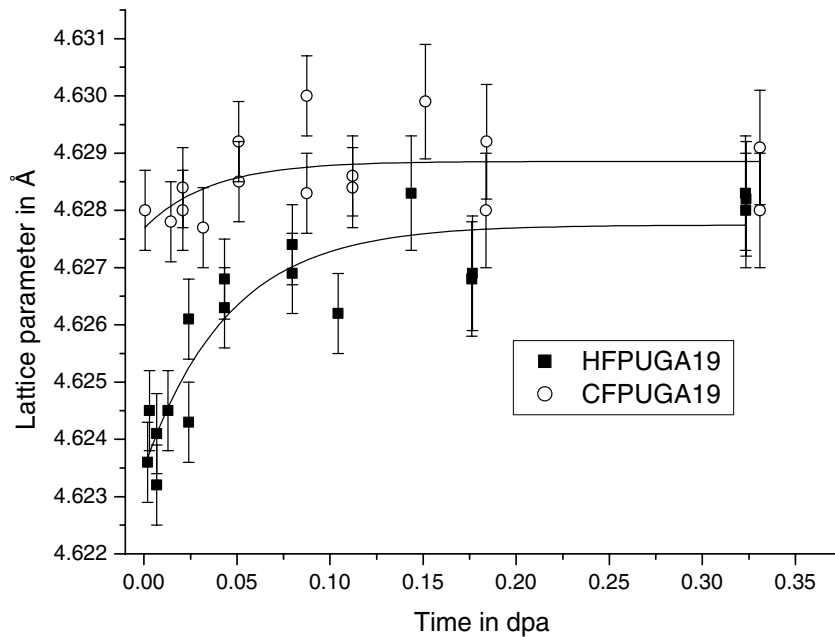

Fig. 3. Change of lattice parameter of fresh cored and homogenized samples (HFPUGA19 and CFPUGA19) of plutonium–gallium 1.9 at.% alloys as a function of time expressed in dpa.
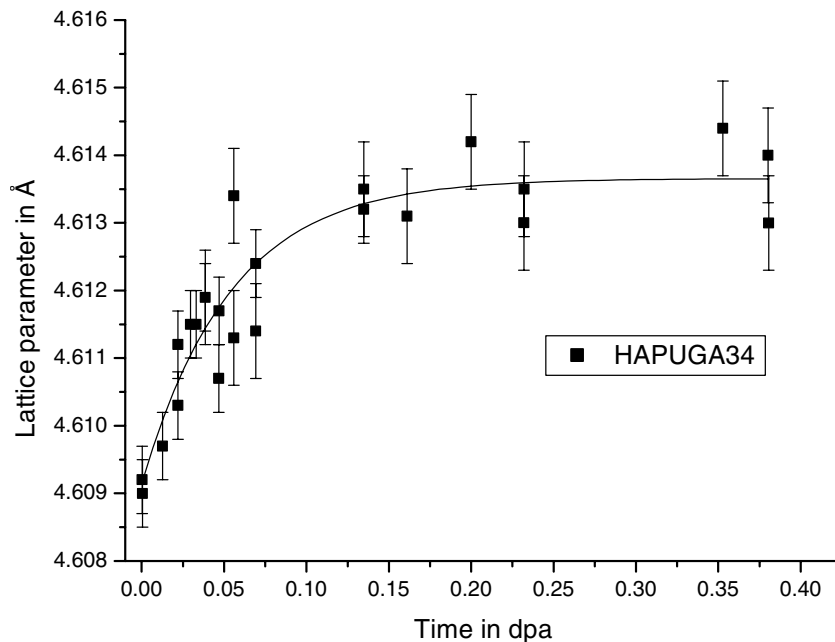

Fig. 4. Change of lattice parameter of aged, heat treated and homogenized sample HAPUGA34 of plutonium–gallium 3.4 at.% alloys as a function of time expressed in dpa.
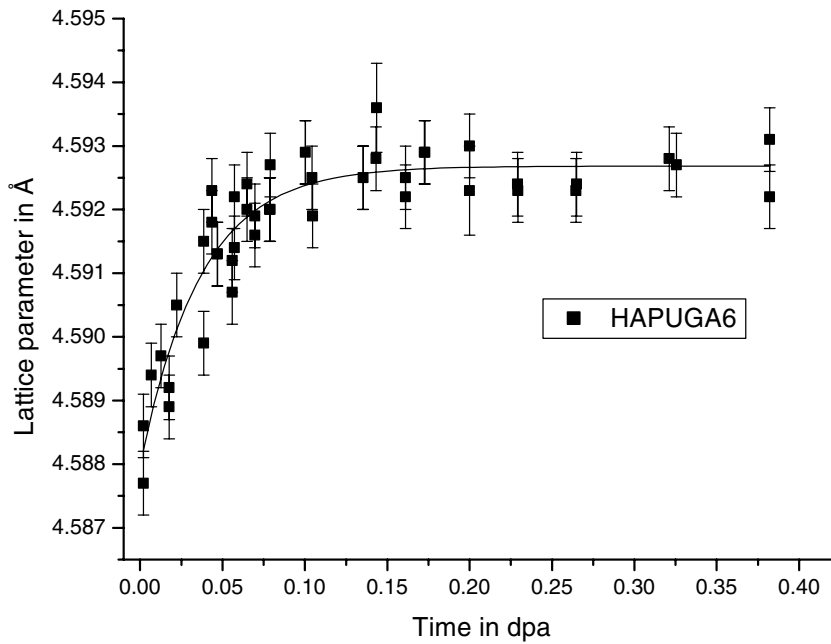
Fig. 5. Change of lattice parameter of aged, heat treated and homogenized sample HAPUGA6 of plutonium–gallium 6 at.% alloys as a function of time expressed in dpa.

for the interpretation of cored plutonium–gallium alloys aging.

3.3. Change of lattice parameter

Figs. 3–5 show the change of lattice parameter versus time for the homogenized samples, HFP-UGA19, HAPUGA34 and HAPUGA6 respectively. These three lattice parameters evolution seem to have the same shape with time (or self-irradiation dose) whatever the gallium content is. More precisely, the lattice parameter quickly increases before reaching saturation for a dose of about 0.1 dpa. The experimental data were fitted using the following mathematical relationship [23–26]:

$$a_{\mathrm{t}} = a_0 + A(1 - \exp(-Bt)), \tag{1}$$

where a_0 is the initial lattice parameter, A the lattice swelling amplitude and B a factor corresponding to the lattice swelling kinetics.

The results of this curve fitting, given in Table 2, show that the gallium content does not affect the swelling amplitude. Indeed, the A factor values are equivalent for all the three homogenized alloys. An increase in lattice parameters close to 4.5×10^{-3} Å is observed at saturation. Likewise, the values of the relative swelling at saturation ($\Delta a/a$) are nearly equivalent. Concerning kinetics of swelling, the B factor values remain close to each other and do not seem to be influenced by the gallium content. The small difference between the studied alloys can be explained by the dispersion of data around the fitted curves.

To study the influence of gallium distribution on the lattice swelling, cored and homogenized samples

Table 2
Parameters determination after fitting using the relationship $a_{\mathrm{t}} = a_0 + A(1 - \mathrm{e}^{-Bt})$ describing the lattice swelling as a function of time for plutonium–gallium alloys

Alloys	a_0 (Å)	A (Å)	B (dpa^{-1})	Lattice parameter at saturation (Å)	Relative swelling at saturation	Relative swelling at saturation (Chebotarev and Utkina [16])
HFPUGA19	$4.6235 \pm 4 \times 10^{-4}$	$(4.2 \pm 0.5) \times 10^{-3}$	22.1 ± 7	$4.6277 \pm 5 \times 10^{-4}$	$(9.1 \pm 2) \times 10^{-4}$	4.0×10^{-4}
CFPUGA19	$4.6277 \pm 6 \times 10^{-4}$	$(1.2 \pm 0.6) \times 10^{-3}$	27.9 ± 32	$4.6289 \pm 6 \times 10^{-4}$	$(2.6 \pm 2.6) \times 10^{-4}$	–
HAPUGA34	$4.6091 \pm 4 \times 10^{-4}$	$(4.5 \pm 0.4) \times 10^{-3}$	18.4 ± 4	$4.6136 \pm 4 \times 10^{-4}$	$(9.8 \pm 1.7) \times 10^{-4}$	7.25×10^{-4}
HAPUGA6	$4.5880 \pm 3 \times 10^{-4}$	$(4.7 \pm 0.3) \times 10^{-3}$	27.3 ± 3.3	$4.5927 \pm 3 \times 10^{-4}$	$(10.3 \pm 1.3) \times 10^{-4}$	13.1×10^{-4}

(CFPUGA19 and HFPUGA19) were characterized by XRD as a function of time just after casting. The evolution of lattice parameter versus time is presented in Fig. 3 for both samples and the data resulting of the curve fitting are given in Table 2. Results show a difference of lattice swelling amplitude of 3×10^{-3} Å, corresponding to the difference between $A_{HFPUGA19}$ and $A_{CFPUGA19}$.

4. Discussion

The measurements of change of lattice parameters for the three studied plutonium–gallium alloys revealed a lattice swelling with no incubation time. This lattice swelling suggests that the defects are randomly distributed and their concentration is sufficient to influence the long range order. As a matter of fact, an increase in the lattice parameter with time may be attributed to an increase in the defects concentration rather than an increase in defect size, which would not affect the lattice parameter. As the samples aged at room temperature, the individual self-interstitials and self-interstitial clusters might not attend since these defects migrate through dislocations or grain boundaries from 20 to 150 K during the stage I and II [14]. Moreover, vacancy activation, vacancy clustering and impurity interactions and finally vacancy clusters dissolution occur within the range 150 and 310 K (stages III, IV and V). Then, at room temperature this suggests that, mainly vacancy clusters contribute to swelling. A combination between helium atoms and vacancies can also occur since helium atoms stabilize vacancies or vacancy clusters [6]. Lattice swelling would then be induced by these vacancy type defects.

Saturation, which is reached for about 0.1 dpa, could then be the result of a competition between production rate of these defects and their recovery, which leads to an equilibrium. Moreover, an overlap of displacement cascades might also occur. This phenomenon can anneal the defects induced by a previous displacement cascade, which would lead to a constant number of defects, i.e. saturation of the lattice swelling. Taking into account the rate of the α-decay of plutonium-239 (about 2.3×10^9 Bq g^{-1}) as well as the observed saturation at 0.1 dpa, the calculated diameter of the displacement cascade could be estimated at 12 nm, which is relatively closed to the value of 7.5 nm previously published by Wolfer [5]. For the three studied homogenized alloys, the saturation of swelling is reached for 0.1 dpa. This value is at least twice fas-

ter than the results reported by Chebotarev and Utkina [16] who claim a saturation after 2.5 years, which corresponds to about 0.25 dpa. The swelling has also been investigated at a macroscopic scale in previous studies. Caturla et al. [27] performed kinetic Monte Carlo simulations to study the evolution of microstructure in irradiated fcc metals with the presence of helium. This modeling showed an increase in swelling at a macroscopic scale. In an other way, the macroscopic swelling has also been experimentally measured by optical fiber Bragg grating dilatometry on a plutonium–gallium 1.9 at.% alloy [28]. During a transient period that lasted until about 0.1 dpa, sample length increased ($\Delta l/l$ $\sim 10^{-4}$ at 0.1 dpa) and then the swelling continued but less rapidly following a linear trend [29]. No saturation is then observed. The origin of the different behavior of macroscopic and microscopic swelling is not fully understood yet even if it might be related to the increase in larger defects number such as helium bubbles.

Regarding the microscopic swelling investigated in the present work, in the studied range of gallium content, the amplitude of lattice swelling is not influenced by the gallium concentration and an average difference of about 4.5×10^{-3} Å is obtained at saturation for homogenized alloys. These results are different from those published by Chebotarev that showed an increase in the relative lattice swelling with the gallium content as shown in Table 2. However, these results can be compared with those obtained on 10 years old plutonium–aluminum alloys with an aluminum content of 11.5 and 12.8 at.% that aged at room temperature [15]. These show a relative lattice swelling of 10^{-3} corresponding to a difference in lattice parameter close to 4.5×10^{-3} Å. A direct comparison can be performed between plutonium–gallium and plutonium–aluminum alloys because the stabilization of the δ-phase is the result of a 5f(Pu) and 3p(Al) or 4p(Ga) hybridization, the configuration of valence electrons for both solutes being equivalent. To understand the absence of gallium influence on the lattice swelling, EXAFS experiments performed at different temperatures by Allen et al. [3] can be considered. This technique allowed to determine the pair-specific correlated-Debye temperature which are 110.7 ± 1.7 K and 202.6 ± 3.7 K for Pu–Pu and Ga–Pu pairs, respectively, in a plutonium–gallium 3.3 at.%. alloy. Because the Debye temperature can be related to a measurement of lattice stiffness, these results indicate that the Ga–Pu bonds are significantly stronger

than the Pu–Pu bonds. Thus, during self-irradiation, the probability to find stable vacancies around plutonium atoms may be statistically more important than around gallium atoms since it is more difficult to break Ga–Pu bonds. Thereby, in the range of gallium content studies, from 1.9 to 6 at.%, the gallium content may not strongly influence the self-irradiation defects.

Regarding the coring effect on the alloys aging, the different swelling amplitude for cored and homogenized alloys tends to prove that either the inhomogeneous gallium distribution might influence the nature and the distribution of the defects in the lattice, which can change the lattice swelling, or the inhomogeneous gallium distribution disturbs the lattice parameter determination by XRD analysis masking the self-irradiation effects as discussed for cored and homogenized alloys before aging.

Regarding the americium effect, previous studies performed by XRD on plutonium–americium alloys [30] highlighted an increase in lattice parameter with increasing americium content, due to both the steric and electronic effects. However, the contribution of americium-241, resulting from the β^- decay of plutonium-241 present in the plutonium–gallium alloys studied is not taken into account. Indeed, after an aging corresponding to 0.2 dpa, an americium content of only 240 appm is generated. That leads to a lattice swelling of 8×10^{-5} Å, negligible compared with the swelling at saturation which is close to 4.5×10^{-3} Å.

The lattice swelling origin may be not completely explained by a steric effect induced by the presence of vacancy type defects. Effectively, a much smaller lattice swelling is observed for aged plutonium–americium alloys with an americium content from 8 up to 20 at.% [29]. In a plutonium–americium alloy, the δ-phase stabilization is induced by a localization of the Pu 5f electrons whereas a 5f(Pu) and 4p(Ga) hybridization is at the origin of the δ-phase stabilization in plutonium–gallium alloys. The difference of lattice swelling under self-irradiation suggests that some electronic effects could eventually contribute to the lattice swelling. Besides, a change of electronic structure during self-irradiation was already observed through magnetic susceptibility measurements performed at low temperature on plutonium–gallium and plutonium–americium alloys [31,32]. Moreover, resonant photoemission experiments performed on a new and aged plutonium–gallium alloy stabilized in the δ-phase suggested differences particularly at the Fermi level

which has a strong 5f character [33]. Thus, a study of the possible modifications versus time of the electronic structure of plutonium–gallium alloys studied in this work should be investigated to complete this structural approach presented here and could help to better understand the lattice swelling.

5. Conclusion

Results of the lattice swelling for plutonium–gallium alloys with different gallium contents have been reported allowing a better description of the aging effects at the crystalline lattice scale. The most important results show no incubation period before the beginning of lattice swelling and in the same time the saturation of lattice swelling occurs for a dose of about 0.1 dpa. Moreover, in the range of gallium concentration studied, the lattice swelling amplitude does not seem to be affected by the gallium content. This has been attributed to a higher probability to find stable vacancies around plutonium atoms than around gallium atoms, suggesting that vacancies would tend to increase the cell volume inducing a microscopic swelling. This last point has to be confirmed through studies of the nature of the defects using transmission electronic microscopy or positron annihilation spectroscopy. Moreover, studies of the nature of the defects would allow to link the microscopic and macroscopic swelling. Indeed, the saturation of the lattice swelling suggests that the evolution of the lattice parameter seems to be not very sensitive to large defects such as cavities or helium bubbles, which could be revealed dominating at the macroscopic scale.

Finally, beyond a structural approach, the influence of the electronic structure will have to be considered to complete this work and to better understand the lattice swelling.

Acknowledgements

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