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Determination of elastic moduli at high temperatures for uranium–vanadium alloy and pure plutonium by an ultrasonic method

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Abstract

Using a contact delay-line ultrasonic device, we measured the elastic properties particularly the shear modulus μ of pure plutonium and uranium–vanadium alloy from ambient temperature to 900 K. Several allotropes of these materials have been detected in the explored temperature range. For plutonium metal, measurements reveal a collapse of $\mu(T)$ of about 50% for the $\alpha \rightarrow \beta$ phase change and a small increase of 0.2% for the $\beta \rightarrow \gamma$ phase change. For uranium–vanadium alloy, we observe a monotonic decrease of about 50% of $\mu(T)$ for the $\alpha \rightarrow \beta$ phase change. In order to include the phase transition to describe the variation of $\mu(T)$, a model has been proposed and applied.

Keywords: Actinide compounds; Material characterization; Elasticity; Ultrasonics; High temperature

1. Introduction

The knowledge of the elastic moduli is necessary in calculations with classical mechanical computer codes, both in the elastic and in the elastoplastic regimes. When a structure is stressed versus temperature, these elastic moduli must be known across the whole range of temperature investigated. For materials which are subjected to high temperatures, most of conventional measurement methods, such as mechanical testings, are unsuitable. An alternative solution is an ultrasonic method [1] which has the advantage to determine the elastic moduli [2] in the extreme elastic limit.

Uranium–vanadium alloy and pure plutonium have been the subject of many previous investigations in different domains: physical properties (corrosion, thermal conductivity, thermodynamics, ...). Indeed with respectively three and six allotropes between room temperature and their melting point, physical properties of these materials vary substantially.

The main intention behind the present experiment was to study particularly the shear modulus μ as a function of the temperature of these two materials and to compare it with the Young modulus and previous measurements.

2. Experimental set-up

The determination of the shear modulus depends on the acoustic velocities, in particularly on the shear velocity, which variation versus temperature can be measured. The principle of the experiment consists in measuring the time of flight of the acoustic wave through the sample as described in the Fig. 1. The elastic-wave velocities determination requires the accurate knowledge of the distance of propagation through the solid versus temperature $l(T)$. This is obtained using the thermal expansion of the material. A furnace is used so that the main heating region is located on the sample up to 1000 K. This latter (cylinder of 4.5–10 mm width and 23 mm diameter) is placed between two wave-guides. At the end of each one, two transducers (respectively, 2.25 MHz for the shear mode and 5 MHz for the longitudinal mode) are connected with the same ultrasonic characteristics. The two wave-guides are used to ensure the propagation of the waves through the sample while the thermal resistance of the wave-guides is sufficient to keep the transducers near room temperature.

3. Results

3.1. Uranium–0.2 wt% vanadium

Uranium, with a melting point of 1405 K has three allotropes: α (below 938 K) is orthorhombic, β (938–1040 K) is tetrag-

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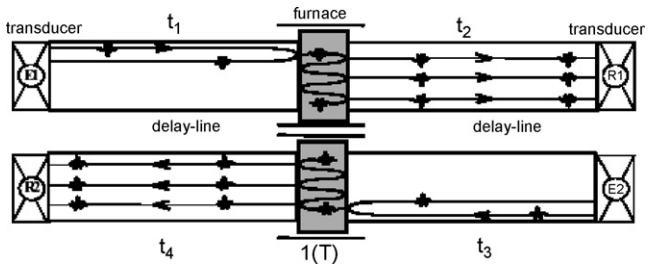

Fig. 1. Principle of measurements.

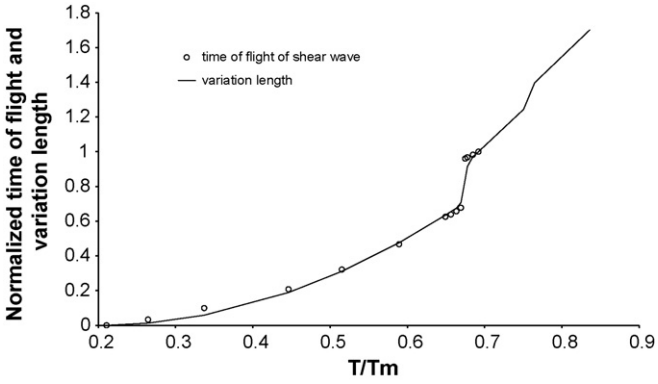

Fig. 2. Normalized time of flight of shear wave and variation length vs. temperature for uranium–0.2 wt% vanadium alloy.

onal and γ (1040–1405 K) is bcc. The addition of vanadium improves mechanical properties of the uranium by reducing the grain size and does not modify the physical properties of the allotropes. Fig. 2 presents the evolution of the normalized time of flight of the shear wave and the normalized variation length of the uranium–0.2 wt% vanadium alloy obtained by [3] versus temperature up to $T/T_m = 0.70$ with $T_m = 1405$ K (T_m: melting temperature). We observed that both ultrasonic and dilatometric data are coherent. One phase transition (α → β) would be noticed and the temperature of phase change is in a good agreement. Evolution of the shear modulus of this alloy has been represented in Fig. 3. The measures have been performed in the temperature range $0.2 < T/T_m < 0.7$ with uncertainties below 4%. The evolution of $\mu(T)$ shows a monotonous decrease of about 50% from its ambient value to the first transition α → β where the fall is drastic (about 20% for a very narrow temperature range).

The results of this study are compared with those of Steinberg [4] for pure uranium. Steinberg et al. proposed the following expression for variation of shear modulus as a function of pressure P and temperature T in the solid state:

$$\mu(P, T) = \mu_{amb} + \frac{\partial \mu}{\partial P}\frac{P}{\eta^{1/3}} + \frac{\partial \mu}{\partial T}(T - 300) \quad (1)$$

where μ_{amb} is the shear modulus at ambient temperature (taken as 300 K), $\partial\mu/\partial P$ and $\partial\mu/\partial T$ are the partial derivatives of the shear modulus versus pressure and temperature, respectively and η is the compression defined by the ratio of the mass density under pressure over the mass density at ambient pressure. In this formula, variation of the shear modulus with the temperature does not depend on pressure.

Assuming linear variation in the shear modulus, results show very good agreement up to the first transition phase. At higher temperature, this model does not take into account phase transition as can be seen in Fig. 3. So in order to describe these phase transitions, we have extended a model based on the description of the solid to liquid drastic fall of $\mu(T)$ [5].

Our model can be written:

$$\frac{\mu(T)}{\mu_{ij}} = \frac{1 + a_{ij}(T - T_{ij})}{\tau(T/T_{ij})}$$

$$\text{for } T \in [0, T_{ij}] \quad (2)$$

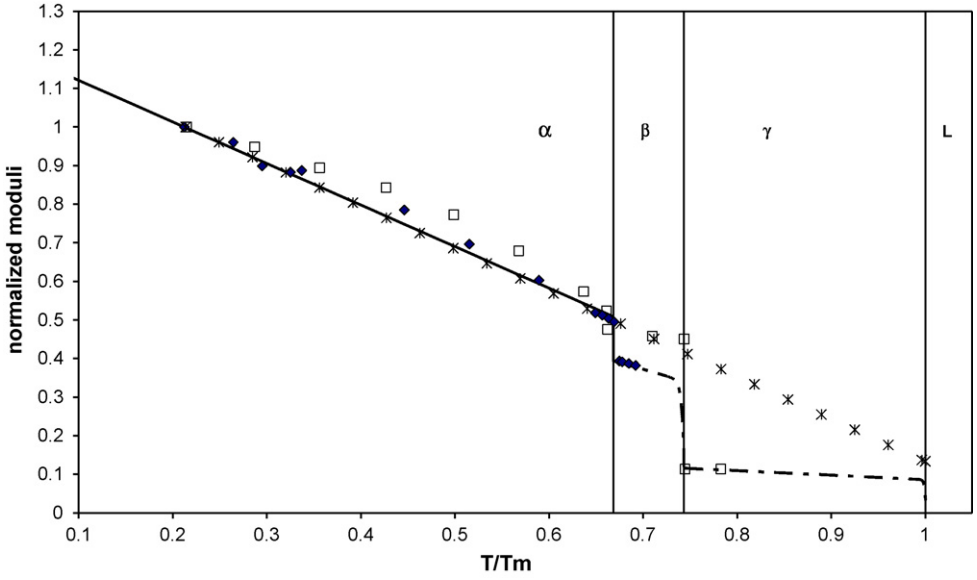

Fig. 3. Normalized $\mu(T)$ of the U–V alloy (♦) with the associated modeling (solid line and dotted line) and $\mu(300\,K) = 86.1$ GPa. Comparison to normalized Young modulus (□) [5] with $E(300\,K) = 201$ GPa and pure uranium Steinberg data (×).

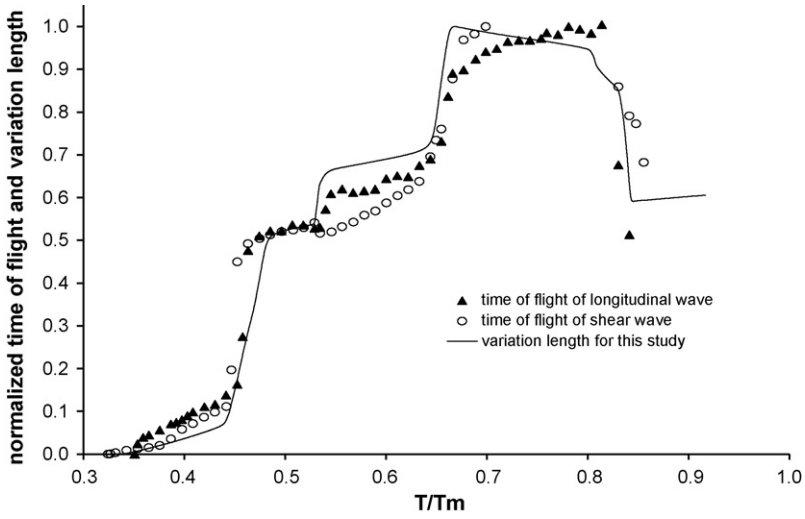

Fig. 4. Normalized time of flight of acoustic waves and variation length vs. temperature for unalloyed plutonium.

$$\tau\left(\frac{T}{T_{ij}}\right) = 1 + \exp\left[\frac{(T/T_{ij}) - 1}{\varepsilon_{ij}\{1 - (T/(T_{ij}(1+\varepsilon_{ij})))\}}\right] \quad (3)$$

where T_{ij} stands for the temperature of transition phase between phases i and j. μ_{ij} is the value of the shear modulus at the temperature T_{ij}. $\mu(T)$ is described by two parameters: a_{ij} (negative slope of the linear variation of μ) and ε_{ij} (coefficient close to 0 which allows the fall of μ at 0 for the temperature just below T_{ij}).

In order to propose a complete description of $\mu(T)$ up to the melting point without anymore experimental data above the $\alpha \rightarrow \beta$ transition, the jump of $\mu(T)$ at $T_{\alpha\beta}$ is supposed to be equal to the Young modulus (E) jump [3]. Then, the $\mu(T)$ evolution up to T_m is proposed.

Table 1 shows the parameters of the model which fit the experimental data and predict the $\mu(T)$ evolution.

3.2. Pure plutonium

Fig. 4 shows the latest results obtained for pure plutonium. The evolution of the normalized time of flight of the acoustic waves and the normalized variation length are compared up to $T/T_m = 0.85$ with $T_m = 913$ K in order to observe the coherence of phase transition and phases jumps. For this comparison, dilatometric measurements have been performed with the same rate of change in temperature used for ultrasonic measurements (2 K/min) on pure plutonium (starting density 19.45 g/cm^3). At the beginning of the experiment, the samples are provided by casting without any quenching. No texture appears, the polycrystalline samples do not exhibit any specific orientation and are considered as isotropic elastic continua.

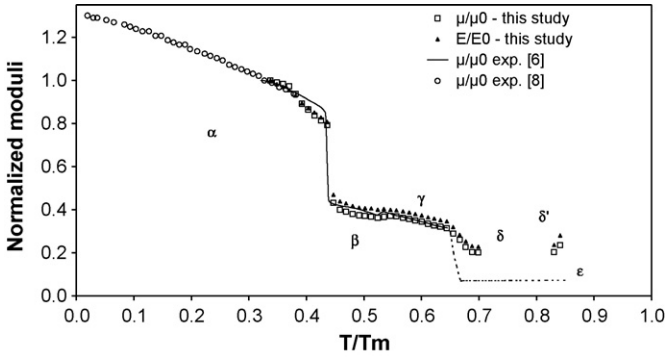

Fig. 5. Normalized $\mu(T)$ and $E(T)$ of unalloyed plutonium with $\mu(300\,\text{K}) = 42.9$ GPa and $E(300\,\text{K}) = 102$ GPa. Comparison to previous Kay data and Migliori data.

We observed that ultrasonic and dilatometric data track each other accurately. Four allotropes (α, β, γ and δ) would be noticed. The temperatures of phase changes are in a good agreement even if a small difference exists around the temperature of $\alpha \rightarrow \beta$ transition.

Evolution of the normalized moduli has been represented in Fig. 5. The results obtained for $T/T_m > 0.7$ are not as accurate as at lower temperatures because beyond this temperature the sample dimensions are not constant due to a probable softening of the metal (δ-phase). The results are compared at room

Table 1
Parameters for model fitting the $\mu(T)$ function

μ_{ij} (GPa)	a_{ij}	T_{ij}	ε_{ij}
$\mu_{\alpha\beta} = 33.9$	$a_{\alpha\beta} = -0.72$	$T_{\alpha\beta} = 938$	$\varepsilon_{\alpha\beta} = 0.01$
$\mu_{\beta\gamma} = 10$	$a_{\beta\gamma} = -1.3$	$T_{\beta\gamma} = 1043$	$\varepsilon_{\beta\gamma} = 0.05$
$\mu_{\gamma f} = 0$	$a_{\gamma f} = -1$	$T_{\gamma f} = 1405$	$\varepsilon_{\gamma f} = 0.03$

Table 2
Ambient measured α-plutonium shear modulus (GPa)

μ (GPa)	Source
42.9	Present
41.5, 34.5 (after 3 thermal cycles)	Kay and Linford [6]
39.1 (at 318 K)	Calder [7]
43.6	Migliori [8]
46	Merz [9]
41.6	Laquer [10]

temperature with previous measurements (see Table 2), below the room temperature with results of Migliori [8] and at higher temperature with those of Kay and Linford [6] which data are obtained by using an ultrasonic resonance method after a fourth cycle of heating the specimen. The regions with large errors, shown as dotted lines in Fig. 5, are included merely to show the range of the heating cycle. We observe that our results are in good agreement with these measurements. The thermal cycling has shown no impact on the variation of $\mu(T)$.

We noticed, like Kay and Linford, an interesting feature shown by both the E and μ graphs (Fig. 5). Indeed there is a small increase of about 0.15% in normalized moduli at the $\beta \rightarrow \gamma$ phase transition change, despite a decrease in density of 3.5 percent. At the $\alpha \rightarrow \beta$ phase change a 10% decrease in density is accompanied by a decrease in both moduli of about 50%.

At the $\gamma \rightarrow \delta$ phase change, we observe a similar behavior with a smaller collapse.

4. Conclusion

This paper shows the first complete shear modulus behavior versus temperature for pure plutonium and uranium–vanadium alloy. For this latter, our measurements have to be completed by longitudinal wave velocity measurements to determine all the elastic moduli and complete the modeling. Concerning the plutonium, additional measurements have to be performed to understand the elastic properties of this material and particularly for the $\beta \rightarrow \gamma$ transition. In a second step, we will test our model developed for uranium–vanadium alloy on plutonium.

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