Experimental investigation of dynamic compression and spallation of Cerium at pressures up to 6 GPa

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Abstract. In this study the experiments on one-dimensional dynamic compression of Cerium (Ce) samples to pressures of 0.5 to 6 GPa using various types of explosively driven generators were conducted. VISAR laser velocimeter was used to obtain Ce free surface velocity profiles. The isentropic compression wave was registered for γ-phase of Ce at pressures lower than 0.76 GPa that corresponds to γ-α phase transition pressure in Ce. Shock rarefaction waves were also registered in several experiments. Both observations were the result of the anomalous compressibility of γ-phase of Ce. On the basis of our experimental results the compression isentrope of Ce γ-phase was constructed. Its comparison with volumetric compression curves allowed to estimate the magnitude of shear stress at dynamic compression conditions for Ce. Spall strength measurements were also conducted for several samples. They showed a strong dependence of the spall strength of Ce on the strain rate.

1. Introduction
Cerium (Ce) has many unusual properties as compared to other metals. Among them is the existence of a solid-state critical point on the line of the isomorphic γ-α phase transition and the anomalous behavior of compressibility, thermal expansion, heat capacity and other properties of γ-phase Ce [1]. Thus, a rather complex pattern is formed on the Ce phase diagram in the region of relatively low pressures (P<25 GPa) [2,3], which in turn results in the appearance of complex multi-wave structures during fast dynamic loading of Ce samples. This makes Ce an important object for studies aimed at constructing multi-phase equations of state of matter. Hence a large volume of experimental and computational effort on the research of various regions of Ce phase diagram [1-4] has been done lately.

In particular, it was shown theoretically in [2] that the anomalous behavior of γ-phase Ce compressibility up to the pressure of γ-α transition lead to the impossibility of shock front formation in that region. It means that compression of Ce must be isentropic there instead. With further increase of pressure an appearance of a two-wave configuration consisting of the head-on wave of isentropic compression which is blurred in time and the following shock wave was expected.

Predicted two-wave structures were actually observed in experiments (e.g. [1,4]). However, the available experimental data still leave a number of questions concerning γ-α phase transition in Ce in dynamic conditions unanswered. Among them are the influence of elastic-plastic properties on the phase transition pressure, the character of compression wave propagation and its structure in γ-phase, and the dependence of the Ce spall strength on the strain rate. This work is dedicated to the experimental investigation of these problems.
2. Scheme of experiments
As the object of experiments, Ce samples with 6.75 g/cc density and 99.83% purity made with electrolysis technique were used. One-dimensional dynamic compression of 1.5, 2 and 4 mm thick Ce samples with 30-50 mm diameter was created by the impact of 0.4-2.0 mm thick metallic flyer plates accelerated with three different types of explosively driven shock-wave generators (see figure 1).

Figure 1. Schemes of experiments for Ce studies using explosively driven shock-wave generators: (a) - compact explosive generator (1 - explosive generator, 2 - 1 mm thick Al flyer plate, 3 - Ce sample, 4 - 7 µm thick Al foil, 5- Steel attenuator plate); (b) - generator of low amplitude shock waves (1 - explosive charge, 2 - Al shield plate, 3 - 50 mm long water-filled container, 4 - Plexiglas plate, 5 - Ce sample, 6 - 7 µm thick Al foil); (c) – “generic” explosive plane-wave generator (1 - plane-wave explosive lens, 2 - Cu attenuator plate, 3 - 0.4-2.0 mm thick Al flyer plate; 4 – 2 mm thick Al shield plate, 5 - Ce sample, 6 - 7 µm thick Al foil).

First experiments were conducted using compact explosive generators of plane shock waves (see figure 1(a)) that had been developed earlier specially for a use in proton radiography studies [5,6]. They allow to accelerate 1-2 mm thick metallic flyer plates to velocities of 0.7 to 2.8 km/s with as little as 10 g of high explosives (HE). Generator of low amplitude shock waves (see figure 1(b)) which is based on the attenuation of a shock wave during its passing through the thick water layer was used to obtain loading pressures lower than the pressure of γ-α phase transition in Ce which is about 0.8 GPa in static compression conditions [2]. Finally, the rest of experiments was conducted in the scheme using “generic” big explosive plane-wave generators [7] (see figure 1(c)) that allowed to accelerate Al flyer plates to 0.7 km/s with about 150 g of HE. Thus, using these three types of explosively driven shock wave generators, dynamic compression pressures of 0.5 to 6 GPa were obtained in Ce samples. Free surface velocity profiles were registered for them with the VISAR laser velocimeter. 7 µm thick Al foils placed at free surfaces of samples were used to reflect probing VISAR laser beams.

3. Results and analysis
A typical free surface velocity profile obtained in one of the experiments with compact explosive generators is shown in figure 2.

Figure 2. Free surface velocity profile obtained in a compact explosive generator experiment.
One can see that the theoretically predicted for $\gamma$-$\alpha$ phase transition in Ce [2] two-wave configuration consisting of the head-on wave of isentropic compression which is blurred in time and the following shock wave is observed in this case. After reaching the maximum value, free surface velocity starts to decrease when the rarefaction wave comes from the direction of the flyer plate. The following velocity oscillations are due to a spall fracture of the sample under tensile stress.

For more detailed study of the vicinity of $\gamma$-$\alpha$ phase transition, explosively driven generators that allowed to reach lower pressures were used. Corresponding experimental results are shown in figure 3. Free surface velocity profiles were obtained for pressures higher (figure 3(a)) and lower (figure 3(b)) than the pressure of phase transition.

![Figure 3](image)

**Figure 3.** Free surface velocity profiles for Ce samples loaded with: (a) 0.4 mm thick Al plate accelerated to 700 m/s with “generic” plane-wave generator (1 - 1.7 mm Ce sample and 2 mm Al shield plate, 2 - 4 mm Ce sample and 2 mm Al shield plate); (b) - generator of low amplitude shock waves (1 - 1.8 mm Ce sample, 2 - 4 mm Ce sample).

At pressures slightly higher than the phase transition pressure the two-wave configuration is still formed (see figure 3(a)). The most interesting observation here is the progressive “blurring” of this configuration during its propagation to a deeper distance within a sample. It is due to the anomalous compressibility of $\gamma$-phase Ce that prevents the appearance of shock waves in it and thus results in isentropic character of compression waves passing through it. It should also lead to a shock nature of rarefaction waves in $\gamma$-phase, and indeed such a shock rarefaction wave is distinctly observed at the profile 1 in figure 3(a).

At a pressure lower than the phase transition pressure the two-wave configuration is absent (see figure 3(b)). However, the blurring of the compression wave front is still observed. It becomes more distinct with the increase of a sample's thickness.

It is important to note that the transition from $\gamma$ to $\alpha$ phase does not correspond to a certain point on a velocity profile (e.g. the inflection point on profile 1 in figure 3(a)), so it is very difficult to determine with good precision the phase transition pressure from the obtained experimental wave profiles. One of the reasons for that is the re-reflection of the first wave as a result of its circulation between a free surface and a shock wave corresponding to $\alpha$ phase. This leads to the appearance of a smooth second wave instead of the expected shock wave.

It is particularly clearly expressed in figure 4. The velocity profile shown there is smoothed due to a circulation of waves. This leads to the disappearance of the salient point corresponding to the phase transition which should be expected on the profile. An approximate location of the salient point can be determined as the intersection point of the extrapolated functions of free surface velocity on time in the regions corresponding to $\gamma$ and $\alpha$ phases. The results of the extrapolation are shown in figure 4 with dashed lines. Their intersection point gives the position of phase transition as 155±5 m/s.

To determine the phase transition pressure, one should take into account the isentropic character of compression in $\gamma$ phase Ce. If the wave front entering a sample can be considered a shock jump, then a
centered compression wave should evolve in these conditions in the same way as centered rarefaction waves are formed in materials with normal compressibility. To prove this, the wave profiles shown in figure 3(a) are redrawn in the $t/h$ coordinates in figure 5, where $t$ is time, $h$ is the thickness of a sample. It is clearly seen that the velocity profiles coincide with each other up to a certain point where rarefaction waves come. It means that the wave flow is indeed self-similar and a compression wave is indeed centered in this case.

**Figure 4.** Determination of phase transition pressure.

**Figure 5.** Free surface velocity profiles for Ce in reduced coordinates: 1 - $h = 1.7$ mm; 2 - $h = 4$ mm.

It allows to determine the compression isentrope of $\gamma$-phase Ce from the obtained experimental data. From the conservation of Riemann invariants [8] follows the condition of conservation of sound velocity along characteristic curves, so these characteristics appear as a batch of straight lines. This allows to calculate the Lagrangian sound velocity $a_L$ along each of the characteristics knowing the exact time of its coming to a free surface $t_f$:

$$a_L = \frac{h}{t_f + h/c_{10}},$$  \hspace{1cm} (1)

where $c_{10}$ is the sound velocity at zero pressure.

As particle velocity $u$ which is equal to half of free surface velocity is known at every moment, one can construct the dependence of $a_L$ on $u$. Knowing the $a_L(u)$ function, one can in turn obtain the compression isentrope in pressure $P$ – specific volume $V$ plane using equations of conservation of Riemann invariants along characteristics [8]:

$$dP = \rho a_L du, \hspace{1cm} dV = du/\rho a_L,$$ \hspace{1cm} (2)

from which $P$ and $V$ can be directly expressed as functions of particle velocity $u$. $P(V)$ isentrope obtained by excluding $u$ from equations is shown in figure 6 as curve 1. It can be seen that the isentrope indeed has negative curvature as it should be for materials with anomalous compressibility.

One should also note that the constructed $\gamma$-phase Ce isentrope is in fact the dependence of longitudinal stress $\sigma_{11}$ on specific volume $V$, because due to elastic properties of Ce its bulk compression curve has to lie lower in figure 6 than the curve 1. A precise position of bulk compression curve cannot be determined from our experiments. However, it can be obtained from measurements of bulk sound velocity on pressure conducted for Ce in [9, 10]. Corresponding results are shown in figure 6 as curves 2 and 3. The difference between the isentrope and the bulk compression curves is due to the shear stress. Its value $\tau = 0.75*(\sigma_{11} - P)$ as a function of deformation $\Delta V/V_0$ is shown in figure 7 for both bulk compression curve estimations 2 and 3. It can be seen that the maximum value of shear stress that depends on dynamic yield stress can reach almost 0.1 GPa.
Figure 6. 1 – Compression isentrope of $\gamma$-phase Ce, 2 – Ce bulk compression curve [9], 3 – Ce bulk compression curve [10].

The obtained shear stress value is comparable with the phase transition pressure and thus influences its magnitude. As stated above, the phase transition happens at the free surface velocity of about 150 m/s, which corresponds to the longitudinal stress of 840 MPa (dashed line in figure 6). In this case the pressure of bulk compression is equal to 760 MPa which fully corresponds to the phase transition pressure in static conditions [10]. Thus, the phase transition pressure remains independent of conditions of deformation, whether it is bulk deformation in static conditions or uniaxial deformation under fast dynamic loading.

Spall strength measurements for Ce were also conducted in addition to the investigation of $\gamma$-$\alpha$ phase transition. As it was mentioned above, the reflection of a compression wave from the free surface of a sample leads to a rise of tensile stress in it which can lead to a spall fracture. Spall strength $P_s$ of Ce samples was calculated for several free surface profiles shown in figure 8 using the equation for elastic-plastic matter [11]:

$$P_s = \rho_0 \frac{c_0 - c_l}{c_0 + c_l} \Delta W,$$

where $c_l = 2.23$ km/s is the longitudinal sound velocity measured with ultrasonic technique; $c_0 = 1.68$ km/s is bulk sound velocity [9]; $\Delta W$ – difference between the maximum and the minimum values on a velocity profile before a spall pulse. The values of spall strength calculated in this way increase almost by a factor of three, from 0.3 to 0.8 GPa, at the increase of the strain rate by an order of magnitude from $1.5 \times 10^4$ to $8.5 \times 10^5$ s$^{-1}$.

Figure 8. Spall strength analysis for free surface velocity profiles obtained in experiments with the following parameters: 1 – 4 mm Ce, 2 mm Al flyer plate; 2 – 1.5 mm Ce, 0.4 mm Al flyer plate; 3 – 2 mm Ce, 0.4 mm Al flyer plate.
4. Conclusion
The experiments on one-dimensional dynamic compression of Cerium showed that $\gamma$-$\alpha$ phase transition leads to the appearance of a two-wave configuration at the amplitude of compression pulse higher than 0.8 GPa. Compression wave in $\gamma$ phase is isentropic due to the anomalous compressibility of Cerium. The compression isentrope is constructed on the basis of experimental data. Its comparison with the bulk compression curve allows to estimate the magnitude of shear stress, which is about 0.1 GPa. As a consequence, the longitudinal stress at the phase transition increases against the bulk compression pressure of 0.76 GPa which is equal to the phase transition pressure obtained in static conditions. It is also shown that the strain rate in the unloading region strongly influences the spall strength of Cerium.

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