PRESSURE DERIVATIVES OF ELASTIC CONSTANTS OF ALUMINUM ALLOY AMg6 AND NANOSTRUCTURED ALLOY n-AMg6. COMPARISON OF EXPERIMENTAL AND CALCULATED FROM THIRD-ORDER ELASTIC CONSTANTS DATA

ПРОИЗВОДНЫЕ ПО ДАВЛЕНИЮ УПРУГИХ ПОСТОЯННЫХ В АЛЮМИНИЕВОМ СПЛАВЕ АМг6 И НАНОСТРУКТУРИРОВАННОМ СПЛАВЕ n-АМг6. СРАВНЕНИЕ ЭКСПЕРИМЕНТАЛЬНЫХ И РАССЧИТАННЫХ ИЗ УПРУГИХ ПОСТОЯННЫХ ТРЕТЬЕГО ПОРЯДКА ДАННЫХ

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Abstract: *Research results for the nonlinear elastic characteristics of the polycrystalline aluminum alloy AMg6 and AMg6/C_{60} nanocomposite (n-AMg6) have been described. The ultrasonic study of the elastic characteristics at pressures up to 1.6 GPa has been carried out using a high-pressure ultrasonic piezometer based on the piston-cylinder device. Pressure dependencies of the longitudinal and shear elastic waves velocities and densities of AMg6 and n-AMg6 were experimentally measured and the pressure derivatives of the second-order elastic constants were calculated. The results derived by this method have been compared with the results of studies of the nonlinear elastic properties of the test alloys using the Thurston–Brugger quasi-static method, where third-order elastic constants have been measured. The relationships between the pressure derivatives of the second-order elastic constants of these alloys and the Mg-content and nanostructuring were established.*

***KEYWORDS**: AMg6 ALLOY, AMg6/C_{60} NANOCOMPOSITE, SECOND- AND THIRD-ORDER ELASTIC CONSTANTS, PRESSURE DERIVATIVES OF THE ELASTIC CONSTANTS.*

1. Introduction

Nonlinear elastic characteristics, such as second-order elastic constants C_{ijkl} (SOECs) and its pressure derivatives, third-order elastic constants C_{ijklqr} (TOECs), are important physical parameters of materials. They describe the anharmonic properties of the crystal lattice, such as thermal expansion, ultrasound attenuation, and efficiency of the interaction of acoustic waves in solids. Nonlinear properties are also important in the nondestructive determination of structural characteristic and mechanical behavior of materials [1,2]. TOECs and pressure derivatives of SOECs are important parameters of the equation of state of materials [3].

Cast alloy AMg6 belongs to the Al–Mg–Mn system, containing 93.68% aluminum, 5.8–6.8% magnesium, and 0.5–0.8% manganese and other impurities [4,5]. It combines good strength and plastic characteristics at room and higher temperatures, and well-welded. This set of properties favored the wide application of this alloy in the aerospace industry, building, and automobile engineering, while the corrosion resistance in various media, including seawater, explains its successful use in shipbuilding. The alloy refers to strain hardening.

In recent, much progress has been made in nanostructuring of solids, leading to new, unusual combinations of properties, such as high strength and ductility combined with high hardness. So, equal channel angular pressing of AMg6 cast alloy allowed one in [6] to obtain the material with the average grain size of 400 nm with the significantly improved mechanical properties (the breaking point σ_B increased from 210 MPa of cast alloy to 440 MPa, and the elongation at break δ is twice augmented to 25% at room temperature). The higher values of mechanical characteristics in nanocomposite carbon-hardened AMg6 alloy prepared via grinding in a ball mill with subsequent extruding were acquired in [7]. The elongation at break in nanocomposite AMg6 was increased to 10.5–14.1% at higher hardness of 1.5–1.7 GPa.

In a series of our studies, we measured the elastic characteristics (SOECs and TOECs) of some of aluminum alloys (B95, D16, AMg6) and the B_4C ceramics [8-10], and investigated the effect of nanostructuring on TOECs values for aluminum alloys n-B95 [11-12] and n-AMg6 [13-14].

In this study we measured the SOECs pressure derivatives for the polycrystalline aluminum alloy AMg6 and the nanostructured n-AMg6 alloy. The experimentally determined TOECs of AMg6 and n-AMg6 alloys [14] are used to evaluate the SOECs pressure derivatives and are compared with the measured values.

2. Linear and nonlinear elastic properties

The linear elastic properties of the solids can be evaluated with the second-order elastic constants C_{ijkl} (SOECs). The measurements of the longitudinal $V_L = \sqrt{C_{11}/\rho}$ and shear $V_T = \sqrt{C_{44}/\rho}$ velocities in bulk acoustic waves (BAWs) and the density ρ_0 of the samples allowed us to determine two of them, the C_{11} and C_{44}, and calculate the Young´s modulus E, the bulk modulus B, the Poisson's coefficient σ and the Lame parameters λ, and $\mu = C_{44}$.

The SOECs for nanomaterials are almost equal to those of the appropriate initial microcrystalline objects. This conclusion is, however, valid for materials with the low interface fraction (the atom content at the interface to the amount of atoms in the bulk). At a crystallite size of less then 10 nm, when this fraction is dozens of percent, the elastic characteristic values are expected to additively reduce owing to the inner porosity in the nanomaterial. Obviously, the pronounced decrease in the elastic characteristics can be avoided by filling the voids of nanoparticle joints with a material possessing high elastic and strength properties (i.e., by forming a nanocomposite), whereas the formation of the chemical bounds between this material and the nanoparticle surface can also favor to the increase in the elastic moduli [15].

To characterize the nonlinear elastic properties of solids the third-order elastic moduli (for isotropic solids) and third-order elastic constants C_{ijklqr} (TOECs) are used. In the isotropic solid, 18 TOECs are nonzero. In this case, three constants C_{111}, C_{112}, and C_{123} are considered as independent; the others are their linear combinations [16]. Also three third-order modules were introduced for isotropic solid. As in the case of second-order modules, the third-order modules can be selected in various ways. Modules ν_1, ν_2, and ν_3 are most commonly used lately [17]. The relationship between them and TOECs is given in [9].

To determine the TOECs, the Thurston–Brugger quasi-static method is used [18]. To do this, relative changes in velocity of bulk waves, depending on the applied uniaxial compression, were experimentally measured in the test samples. In this case, changes in the transit times of ultrasonic waves in the samples are measured as a function of the applied stress P and the derivatives $[\partial (\rho_0 W^2)/\partial P]_{p=0}$ are calculated. From the data obtained, TOECs are determined:

$$[\partial (\rho_0 W^2)/\partial P]_{P=0} = (2 \rho_0 W^2 F + G) \qquad (1)$$

where $W = l_0/t\ (P)$ is the “natural” velocity of BAW, ρ_0 is the density, $F = S_{jkab} M_a M_b U_j U_k$, $G = S_{ipab} C_{jrksip} U_j U_k N_r N_s M_a M_b$, S_{ipab} are the second-order compliance coefficients, U_k is the components of the polarization vector **U**, N_r are the components of the wave vector **N**, and M_i are the components of the unit vector **M** in the uniaxial compression direction.

The experiments on hydrostatic compression directly measure the pressure derivatives of SOECs C_{11} and C_{44}. For the calculation of 3 independent TOECs the measurements in hydrostatic conditions must be supplemented by measurements under conditions of uniaxial compression. However, to determine the three independent TOECs C_{111}, C_{112}, C_{123} in an isotropic solid, it is sufficient to carry out three independent measurements of the dependence of the BAW velocity on the uniaxial compression **P** with the following mutual arrangement of the unit vectors M, N, U:

$\mathbf{N} \parallel \mathbf{U} \perp \mathbf{M}$, $\mathbf{N} \perp \mathbf{U} \parallel \mathbf{M}$, $\mathbf{N} \perp \mathbf{U} \perp \mathbf{M}$.

The direction of uniaxial compression for all measurements must be perpendicular to the propagation direction of the elastic wave. The convolution for ρ_0, W^2, F, G in (1) for these cases is given in [9].

To increase the accuracy of measurements, they are often carried out for more than three combinations of directions of the unit vectors **M**, **N**, **U**.

Third order elastic modules and TOECs of aluminum alloys were studied by many authors for a long time [3].

In [9,13,14], linear and nonlinear elastic properties of samples of a polycrystalline industrial aluminum alloy AMg6, corresponding to GOST 4784-74 and a nanostructured n-AMg6 alloy obtained from it, were studied in detail. The results of these measurements are given in Table 1. The error value of TOECs in Tabl. 1 does not exceed 15%.

***Table 1** Average values of TOECs of AMg6 and nanostructured n-AMg6 (in GPa).*

Material	C_{111} (± 240)	C_{112} (± 120)	C_{123} (± 70)	$\nu_1=C_{123}$ (± 70)	$\nu_2=C_{144}$ (±20)	$\nu_3=C_{456}$ (±10)
AMg6	-3420	-1310	-860	-860	-230	-150
n-AMg6	-2770	-980	-680	-680	-150	-150

3. SOECs pressure derivatives

In the present work, using the data for the TOECs, we calculated the pressure derivatives of the SOECs. The results of calculations are presented in Table 2.

***Table 2** Average values of SOECs pressure derivatives of AMg6 and n-AMg6.*

Material	$\partial C_{11}/\partial P$	$\partial B/\partial P$	$\partial C_{44}/\partial P$
AMg6	27.4	20.7	5.0
n-AMg6	22.2	16.7	4.1

We note that among the known literature data on the SOECs pressure derivative values for aluminum alloys [3,19,20], the largest values are for those containing Mg [3].

The calculated values of the SOECs pressure derivatives for the polycrystalline aluminum alloy AMg6 and the nanostructured n-AMg6 alloy noticeably exceed the values of the literature and tabular data for other aluminum alloys that are close in composition to AMg6.

In order to verify our results of measurements of the TOECs, we conducted direct measurements of the pressure derivatives of the SOECs under conditions of quasi-hydrostatic pressure up to 1.6 GPa.

4. Materials and Experiment

Nanostuctured samples n-AMg6 were prepared from a commercial polycrystalline alloy by refinement and homogenization a mixture of small chips of the alloy with the addition of 0.3 wt.% fullerite C_{60} in a planetary mill AGO-3Y in-cycle work 60 min. The resulting product consists of 200-500 micron agglomerates of nanoparticles. The coherent scattering length (CSL) distribution in powdered samples showed the mean nanoparticle size ~ 40–60 nm.

The process was conducted in a protected Ar-atmosphere; additional studies confirmed the absence of oxygen and the corresponding insertion of aluminum oxidation.

Then, the milled nanopowder was pre-compacted in a cylindrical mold with a diameter of 180 mm at a temperature of 250 ^{0}C and a pressure of 200–300 MPa.

The resulting compact (preform) was subjected to extruding at a temperature of 300 ^{0}C with a reduction of cross-sectional area at least 4 times (in this case to a diameter of 90 mm). The recrystallization process is hindered by grain boundary modification with fullerite C_{60}; the latter plays the role of compacted samples’ stabilizer. A general view of extruded billets and prepared samples for mechanical testing is shown in Fig. 1.

A) B)

***Figure1** General view of the samples n-AMg6/C_{60}:*
A) *after extrusion;* B) *prepared test specimens*

For experiments under hydrostatic compression, the samples in the form of a disk with a diameter of ~16 mm and a height of ~6 mm were cut from the parent AMg6 and nanostructured n-AMg6.

The ultrasonic study of the pressure dependencies of elastic characteristics was carried out using a high-pressure ultrasonic piezometer based on the piston-cylinder device with the registration system of the transmitted and reflected sound built on the basis of the National Instruments PXI platform (Fig.2).

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Changes in the ultrasonic signal paths were measured with 0.005 mm uncertainty using dial-type micrometer indicators. The volume of the samples under pressure was initially determined from the change in their length, and subsequently the equation of state was calculated by the recursive (with respect to density) integration of compressibility (reverse bulk modulus) with accuracy of the order of several %. The measurements of the changes in the transit times (to ±0.001 μs) of longitudinal and transverse ultrasonic pulses were performed by the pulsed method using x-cut and y-cut LiNbO3 plates as piezoelectric sensors with carrier frequencies of 5MHz for transverse geometry and of 10 MHz for longitudinal one. The transit time was measured not only by time shift of the passed sound wave but also by the cross-correlation and autocorrelation method, when one could determine the absolute transit time of the sound wave in a sample. The estimated pressure uncertainty was 0.03 GPa [21].

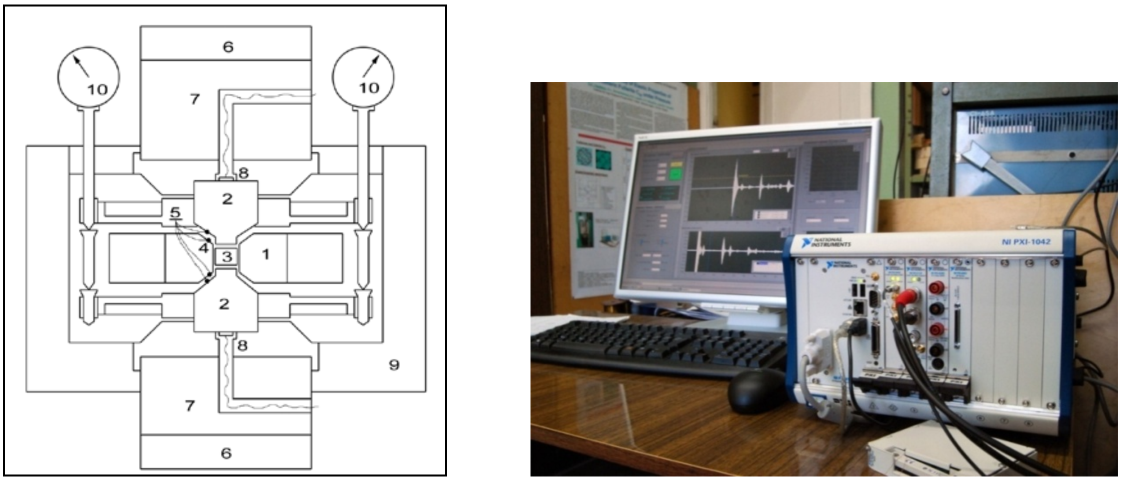
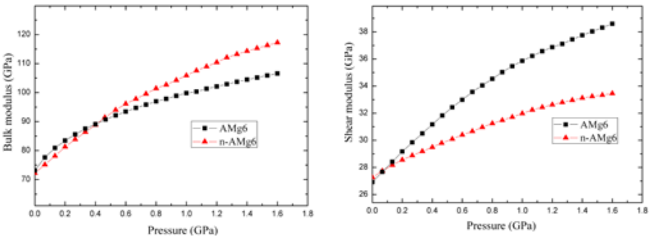


***Figure 2** High-pressure ultrasonic piezometer with the registration system built on the basis of the National Instruments PXI platform. 1 - high pressure cell, 2 - pistons, 3 - sample, 4 - fine copper foil, 5 – thermopairs, 6 - thermal isolation disk, 7 - steel tower, 8 - piesoelectric plates, 9 - thermal isolation tank, 10 - indicating gage.*

Corrections for the deformation of the chamber with the variation of pressure and temperature were determined in special experiments.

5. Results and Discussion

The obtained dependences of the elasticity characteristics on the pressure are shown in Fig.3. First of all, we note the pronounced nonlinear character of the dependence of the moduli on the pressure. Probably, this is due to the peculiarities of pressure generation in the chamber at the beginning of the range.



A) B)

***Figure 3** Pressure dependence of the elastic moduli of AMg6 and n-AMg6: A) B(P), B) G(P)*

In pressure ranges 0.2–0.8 GPa, ∂B/∂P is 26.7 and 33.6 for AMg6 and n-AMg6, respectively, but in pressure ranges 0.8–1.6 GPa ∂B/∂P is 11.3 and 18.9 for AMg6 and n-AMg6, respectively. Values ∂G/∂P in the indicated pressure ranges is 8.9 and 4.5 and 4.6 and 2.5 for AMg6 and n-AMg6, respectively.

In Table. 3 shows the average values of the pressure derivatives. The values for the range 0.8-1.6 GPa are shown in brackets.

***Table 3** Average values of SOECs pressure derivatives of AMg6 and nanostructured AMg6.*

Alloy	Mg(%)	$\partial C_{11}/\partial P$	∂B/∂P	$\partial C_{44}/\partial P$
AMg6[14]	5.8-6.8	27.4	20.7	5.0
AMg6 (Exp.)	7.08	25.1(18.8)	16.0(12.0)	6.8(5.1)
n-AMg6[14]	6.31	22	16.7	4.1
n-AMg6 (Exp.)	6.31	30.7(23.4)	25.8 (19.7)	3.6 (2.8)
D54S[19]	4.5	18.4	12.8	3.5
B53S[19]	2.8	9.7	6.0	2.1

The values of the pressure derivatives turned out to be significantly larger than those known from the literature for aluminum alloys [3]. According to the literature, it is clear that the Mg-containing aluminum alloys have the highest values of the SOECs pressure derivatives, in comparison with others.

In [19], the pressure derivatives of aluminum alloys D54S (Mg-4.5%, Mn-0,8%, Cr-0.1%) and for B53S (Mg-2.8%, Mn-0,8%, Cr-0.1%) were measured. Comparison with these data indicates the presence of dependence of SOECs pressure derivatives on the Mg-content. Our study shown that SOECs pressure derivatives of aluminum alloys belonging to the Al–Mg system increase monotonically with increasing content of Mg in the alloy.

The SOECs pressure derivatives calculated by TOECs and measured in this work agree well with each other with the exception of the ∂B/∂P. According to the data computed from the TOECs values, the SOECs pressure derivatives of the n-AMg6 alloy are reduced. In our experiments, the values of the ∂B/∂P getting larger when nanostructuring. But the values of ∂B/∂P and $\partial C_{11}/\partial P$ measured in the pressure range 0.8-1.6 GPa are in good agreement with those calculated from the TOECs for both AMg6 and n-AMg6.

This can be explained as follows. In [14], the TOEC was measured at uniaxial pressure from 3 to 6 MPa. As can be seen from Fig.3, at a low hydrostatic pressure (< 0.2 GPa), the ∂B/∂P for n-AMg6 is smaller than for AMg6.

6. Conclusion

The pressure derivatives of the second-order elastic constants of AMg6 and n-AMg6 were experimentally determined using a high-pressure ultrasonic piezometer based on the piston-cylinder device. Pressure dependencies of the longitudinal and shear elastic waves velocities and densities and constants were measured at pressures up to 1.6 GPa. The results derived by this method have been compared with the results of studies of the nonlinear elastic properties of the test alloys using the Thurston–Brugger quasi-static method, where third-order elastic constants have been measured. There is a good agreement of the pressure derivatives, obtained by direct measurements under hydrostatic pressure and calculated from TOECs defined by uniaxial loading method.

It is established that nanostructuring leads to a decrease in values of pressure derivatives of all the SOECs.

The values of the pressure derivatives of the SOECs increase monotonically with increasing content of Mg in the alloy.

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