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BULK MODULUS AS A MATERIAL CHARACTERISTIC INDEPENDENT OF TEXTURE

MODUŁ SPRĘŻYSTOŚCI OBJĘTOŚCIOWEJ JAKO CHARAKTERYSTKA MATERIAŁOWA NIEZALEŻNA OD TEKSTURY

It was shown, using as an example a stoichiometric NiAl intermetallic compound of a cubic B2 structure, that the bulk modulus, as an invariant of the tensor of elasticity is a physical quantity describing the properties of a porous polycrystalline material independently of its texture. To show this, the tensor of elasticity for materials of different texture and porosity resulting from different processing routes was measured using ultrasonic method. Variety of texture and porosity in materials were achieved by combination of the following technological processes: casting, self-sustained high temperature synthesis, hydroextrusion and hot isostatic pressing. It was also shown that the value of the bulk modulus is related to the porosity of the material more explicitly than the mean value of Young's modulus.

Keywords: elastic properties, texture, porosity, intermetallic NiAl, ultrasonics

Na przykładzie stechiometrycznego związku międzymetalicznego NiAl o strukturze regularnej B2, pokazano, że moduł sprężystości objętościowej, jako niezmiennik tensora sprężystości, jest dla porowatego materiału polikrystalicznego wielkością niezależną od tekstury. W tym celu, metodami ultradźwiękowymi zmierzono tensor sprężystości materiałów o różnej teksturze i porowatości wytworzonych kilkoma technologiami. Zmiany tekstury i porowatości osiągnięto poprzez kombinacje następujących procesów technologicznych: odlewanie, samopodtrzymująca się synteza wysokotemperaturowa, hydroekstruzja, izostatyczne prasowanie na gorąco. Pokazano także, że wartość modułu sprężystości objętościowej wyraźniej zależy od porowatości materiału niż średnia wartość modułu Younga.

1. Introduction

Elastic properties of any polycrystalline material depend on properties of all phase components of the material and on their microstructure, that is, on the arrangement of these components. In the case of a single-phase material, the main feature of the material is the distribution of crystallographic orientations of grains, i.e. texture. For such a single-phase material direct relations can be theoretically established between the texture and elastic properties of the material when the components of the tensor of elasticity, C_{ij} , for a single crystal are known. These relations can be achieved comparatively easily for the crystal structure with high symmetry, like a cubic one, which is the crystal structure of the majority of metals. Materials possessing the strong elastic anisotropy of their crystals would be especially suitable for experimental verification of such relations.

Elastic anisotropy is completely described by the tensor of elasticity of the crystal but, for simplicity, sev-

eral coefficients of anisotropy are used, like the coefficient of linear compressibility, A_l , and the coefficient of shear stiffness, A_t . For crystals of a cubic symmetry the coefficient A_l is equal to 1 because the linear compressibility of the cubic crystal is independent of the direction [1], but A_t defined as:

$$A_t = \frac{2C_{44}}{C_{11} - C_{12}} \quad (1)$$

can vary in the wide range of values from nearly 1 for tungsten, molybdenum and aluminium up to 7-9 for potassium, sodium and lithium.

The problem becomes more complicated when the material is a mixture of two or more phases. The simplest example of such a material is a single-phase material containing some porosity where the pores can be considered as a second phase with no elastic stiffness. This is a fundamental problem for a wide class of structural materials, both metals and ceramics, but particularly for

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brittle ceramics and semibrittle intermetallics produced via powder technology. They usually contain numerous irregular pores. The problem has been extensively studied both theoretically and experimentally. The review of these works can be found in the literature, for example [2-8]. However, in our opinion, there is still a lack of complete results for materials produced with the use of plastic working, which is often applied for densifying or/and shaping the products. Dependently on parameters of deformation both the various crystallographic texture of the matrix and various morphology of pores can be created. In such a case the assumption concerning isotropy of the matrix can result in misleading conclusions.

The aim of this work was establishing an experimental relationship between the texture and elastic properties of anisotropic and porous material which was a semibrittle intermetallic NiAl with the cubic B2 crystal structure of a stoichiometric composition being regarded as a potential structural material for high temperature applications. Due to strongly directional covalent bonding between nickel and aluminium atoms, the NiAl is highly anisotropic with the coefficient of anisotropy, A_r , equal to 3.74 [9], what results in anisotropy of the final products strongly affecting their mechanical properties.

2. Material preparation

Elemental powders of aluminium of the purity >99.9% (average particle size $\sim 5 \mu\text{m}$) and carbonyl nickel of the purity >99.7% (average particle size $\sim 3 \mu\text{m}$) were used as the raw materials. The main impurity of the nickel powder was carbon ($\sim 0.2\%$) precipitated as fine graphite flakes. Two production routes of NiAl were applied: casting into ingots and powder metallurgy. At the beginning of the first route the rod-shaped compacts were prepared from the powders by cold isostatic pressing. After melting them in a graphite crucible in an induction vacuum furnace the alloy was poured off into the cylindrical graphite moulds of 50 mm in diameter and 300 mm long. For the second route, a stoichiometric composition of powders (50Ni-50Al) was mixed using the steel balls in a cylindrical diagonally rotating mixer. The mixture was compacted into rods of 40 mm in diameter by cold isostatic pressing at the pressure of $\sim 0.3 \text{ GPa}$, sealed in the thick-wall cylinder then heated to the temperature of $\sim 600^\circ\text{C}$ to initiate a strongly exothermic self-sustained high temperature synthesis (SHS) giving a sintered material. Materials obtained via both routes were single-phase ones, what was confirmed by X-ray diffractometry, but porous. To densify, both materials were subjected to plastic working in some consecutive operations by hot hydrostatic extrusion (HE) at

a total reduction ratio of 4, 16 and 64 and temperature $600\text{-}1000^\circ\text{C}$. Then, some samples were densified to nearly theoretical density by hot isostatic pressing (HIP) in an argon atmosphere at the temperature ranging from 900 to 1200°C and pressure 1 GPa . To produce a reference isotropic material of high density some samples of SHS material were directly densified by HIP without any previous HE.

In sum, samples after following combinations of processes were subjected to further elastic constants measurements: 1) cast + HE, 2) SHS, 3) SHS + HIP, 4) SHS + HE and 5) SHS + HE + HIP. The routes 2) and 3) produced isotropic materials while the other ones – textured. In variants with HE: for the route 1 the total reduction ratio was 4, 16 and 64 while for the routes 4 and 5 only 4.

3. Experimental procedure

The samples of the materials were inspected on longitudinal sections of products by a light microscope. The inspection revealed highly inhomogeneous grain size and shape in the as-cast sample and that is why this sample was not subjected to elastic constants measurements. The density of the samples was measured with an accuracy $\sim 0.1\%$ by the Archimedes' principle using water as an immersion medium. The texture was measured by the Schulz back reflection method on specimens' transverse sections.

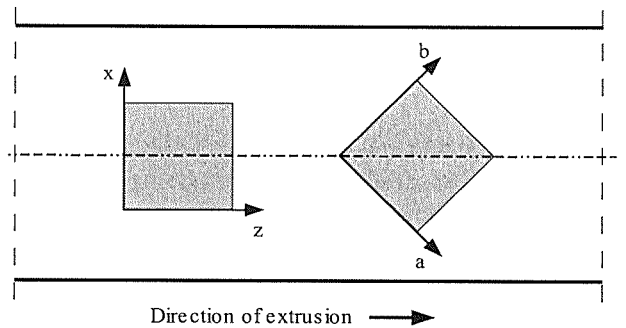


Fig. 1. Orientations of specimens for elastic properties measurements

Elastic properties were determined by an ultrasonic method. The velocities of longitudinal and transverse ultrasonic waves were measured on specimens cut out from the material in the form of parallelepipeds oriented along the extruded rods and at the angle of 45° to the axis (Fig. 1). The dimensions of the specimens were ranging from $\sim 4 \text{ mm}$ for the material after the highest deformation to $\sim 12 \text{ mm}$ for the SHS material. Parallel side walls of specimens were polished with the accuracy of $1 \mu\text{m}$. The waves were generated by the piezoelectric transducers of the frequency of 5 MHz made of lithium

niobate. The transducers were bonded to the waveguides made of beryllium bronze. The time of the wave passing through the sample was measured with the accuracy of 1 ns using a digital oscilloscope with the high frequency of sampling (5 Gs/s). The time of the first measurable extreme on the sinusoidal signal was taken as the reference time for all measurements.

4. Results and discussion

The relative porosity of as-cast material was ~ 0.02 and that of SHS one ~ 0.1 . In both cases the pores had irregular but nearly equiaxed shapes (Fig. 2). The difference in morphology of pores between as-cast and as-sintered materials was quantitative rather than qualitative. This is because, in fact, the sintered material also crystallises from the liquid phase, since the temperature during the SHS process reaches 1800°C [10] that is higher than the melting point of NiAl. Through the HE or HIP the porosity can be reduced even to zero. During HE, the pores become strongly elongated, but the oppo-

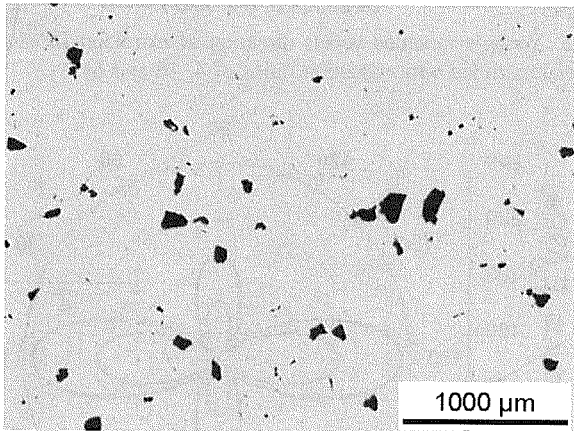


Fig. 2. Optical microphotograph of the material after SHS showing porosity

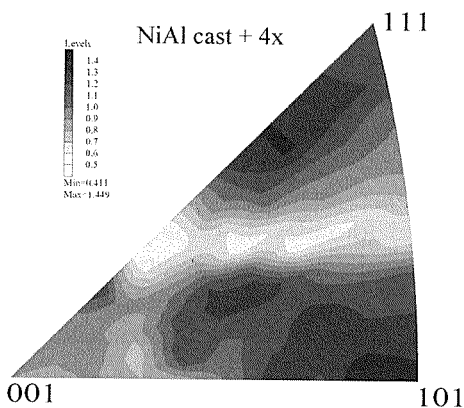


Fig. 3. Inverse pole figure for extruded cast NiAl with a reduction ratio of 4

site surfaces of pores are joined only partially by this process. The texture of both cast and SHS materials is presented in the form of inverse pole figures in Fig. 3 and Fig. 4, respectively. While the SHS material has a sharp texture of the $\langle 110 \rangle$ type, the cast one has a mixed texture of the $\langle 110 \rangle$ and $\langle 111 \rangle$ types. This difference can result from the higher porosity in the SHS material damping the process of dynamic recrystallisation.

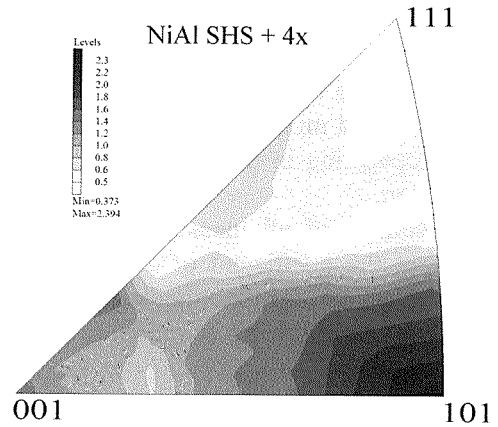


Fig. 4. Inverse pole figure for extruded SHS NiAl with a reduction ratio of 4

In the case of isotropic material the following expressions were used to calculate the values of bulk modulus, B , and Young's modulus, E :

$$B = \rho \left(v_L^2 - \frac{4}{3} v_T^2 \right) \quad (2)$$

$$E = \frac{3B}{A - 1}, \quad (3)$$

where: ρ is the density of the material, v_L and v_T are velocities of ultrasonic waves, longitudinal and transverse, respectively, and

$$A = \frac{v_L^2}{v_T^2}. \quad (4)$$

To calculate the elastic moduli of extruded materials the formulas for transverse isotropy (monotropy) was applied assuming radial homogeneity of the material. There are five elastic constants describing elastic properties of such a material. They were calculated using the following expressions:

$$C_{11} = \rho v_{yy}^2 \quad (5)$$

$$C_{33} = \rho v_{zz}^2 \quad (6)$$

$$C_{12} = C_{11} - 2\rho v_{yx}^2 \quad (7)$$

$$C_{44} = \rho v_{zy}^2 = \rho v_{yz}^2 = \rho v_{zx}^2 \quad (8)$$

$$C_{13} = \left[\rho^2 (v_{aa}^2 - v_{ab}^2)^2 - \frac{1}{4} (C_{33} - C_{11})^2 \right]^{1/2} - C_{44} \quad (9)$$

In these expressions all numeric suffixes of elastic constants are in agreement with the normal shorter tensor notation [1] and they correspond to the notation shown in Fig. 1 as follows: $x \equiv 1, y \equiv 2, z \equiv 3$. The same indices in velocity v_{ii} denote the longitudinal wave propagating in the i direction and mixed indices v_{ij} denote the transverse wave propagating in the i direction and polarized in the j direction.

The correctness of all measurements was verified by the value of the coefficient of anisotropy of linear compressibility, which, for the monotropic material, can be expressed by the following equation:

$$A_l = \frac{C_{11} + C_{12} - 2C_{13}}{C_{33} - C_{13}} \quad (10)$$

Since for cubic crystals A_l is equal to 1, any polycrystalline aggregate of such crystals should possess the same property. In fact, in all our measurements A_l was nearly 1. Using C_{ij} values both bulk and Young's moduli could be calculated. It should be noticed, however, that the bulk modulus expressed by the following equation:

$$B = \frac{(C_{11} + C_{12})C_{33} - 2C_{13}^2}{C_{11} + C_{12} + 2C_{33} - 4C_{13}} \quad (11)$$

is a scalar physical quantity for the monotropic material, while the Young's modulus is a quantity dependent on the direction and can be expressed in the polar coordinates as a function of the angle θ measured from the z axis, i.e. the axis of extrusion (Fig. 1), as follows:

$$E = \left[\frac{C_{11}C_{33} - C_{13}^2}{(C_{11} - C_{12})C} \sin^4 \theta + \frac{C_{11} + C_{12}}{C} \cos^4 \theta + \left(\frac{1}{C_{44}} - \frac{2C_{13}}{C} \right) \sin^2 \theta \cos^2 \theta \right]^{-1} \quad (12)$$

where

$$C = (C_{11} + C_{12})C_{33} - 2C_{13}^2 \quad (13)$$

The dependence of E upon direction defined by θ is presented on polar graphs for representative specimens: for the cast material (Fig. 5) and for the SHS one (Fig. 6). The substantial difference in texture is visible between the cast and SHS materials, what is reflected in the main features of pole figures (Fig. 3 and 4). The first one is less anisotropic. Their elastic anisotropy evolves during

the deformation. The second one is highly anisotropic. This anisotropy was not decreased by HIP (10 h) at the temperature up to 1200°C. From the strong dependence of $E(\theta)$ on the processing route (Fig. 5 and 6) it seems obvious that the attempt of correlation of the Young's modulus (measured in any direction) with the porosity (density) of the material has no sense because of huge scattering of the results.

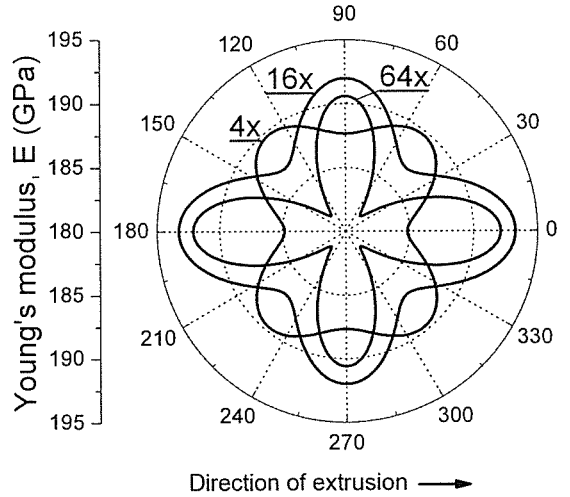


Fig. 5. Young's modulus versus direction of extrusion for the cast material extruded with reduction ratios of 4, 16 and 64

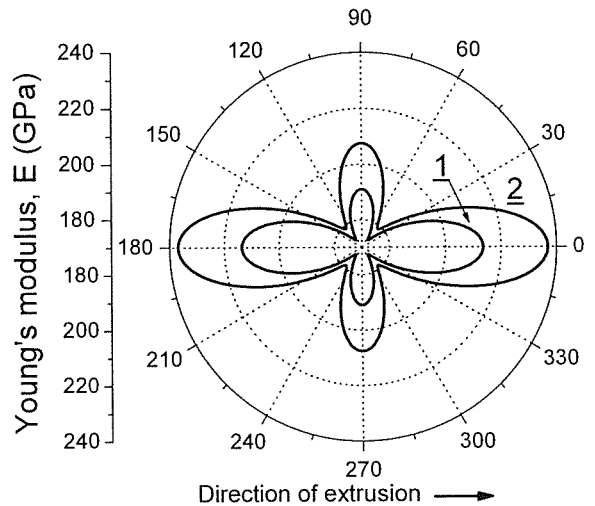


Fig. 6. Young's modulus versus direction of extrusion for the extruded SHS material before – 1 and after – 2 hot isostatic pressing at 900°C

For isotropic materials numerous empirical and semi-empirical formulas describing the dependence of the Young's modulus upon porosity in the form of the combination of polynomial and exponential functions were used to approximate the experimental results [3]. In the case of approximately isotropic, sintered from a ground powder, NiAl the following expression was successfully applied [11]:

$$E = E_0 (1 - \alpha\pi)^n \quad (14)$$

where π is the porosity, a and n are material constants, and E_0 is the value of E for $\pi = 0$, i.e. for the material without pores. An analogous expression can be applied for $B(\pi)$ or $G(\pi)$, where G is a shear modulus.

To describe the effect of porosity on elastic properties of materials with a different type of texture we used two scalar values representing these properties: the bulk modulus, B , and the mean value of the Young's modulus, E_m , which can be determined from an integral of the function $E(\theta)$ (Eq. 12) over all directions of the space. Both values were presented in relation to density, ρ , and porosity, π , in (Fig. 7) and (Fig. 8), respectively.

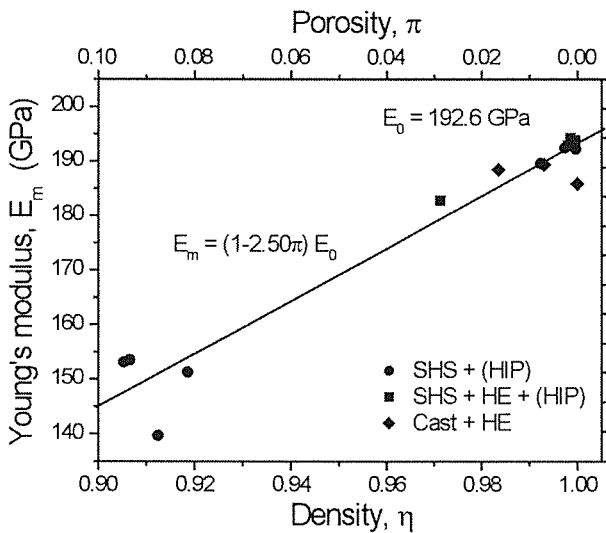


Fig. 7. Mean value of the Young's modulus versus porosity (density); HE – hydrostatic extrusion, HIP – hot isostatic pressing

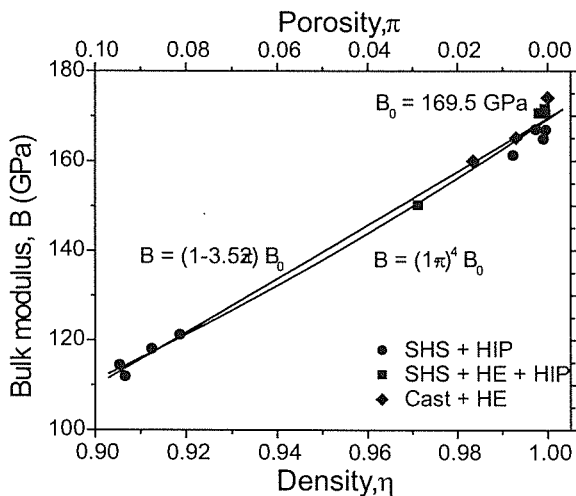


Fig. 8. Bulk modulus versus porosity (density); HE – hydrostatic extrusion, HIP – hot isostatic pressing

The dependence of these scalar quantities upon porosity can be approximated by linear functions:

$$E_m = (1 - a\pi) E_0 \quad (15)$$

$$B = (1 - a\pi) B_0. \quad (16)$$

The first one is fitted with a smaller coefficient of correlation (0.95) because of scattering results caused, in our opinion, by the difference in the shapes of pores, what, in the light of earlier investigations [5,6], has a strong effect on the value of Young's modulus. At the same time the linear approximation of $B(\pi)$ shows a much better coefficient of correlation (0.99), in spite of the fact that it is more sensitive to porosity (the parameter a equals 2.50 and 3.52, respectively).

The dependence of bulk modulus on porosity can be also equivalently well approximated by the expression:

$$B = (1 - \pi)^n B_0 \quad (17)$$

using a nonlinear least square fitting algorithm.

5. Conclusions

It was experimentally confirmed that the bulk modulus of porous and textured polycrystalline materials is a scalar quantity practically independent of texture. Hence, it can be used for separation the effects of porosity and texture on elastic properties of porous and textured materials.

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