Regularities of the influence of temperature and pressure on the grain size in the synthesized intermetallic compound Ni$_3$Al

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Abstract. The intermetallic compound Ni$_3$Al ($\gamma'$-phase, ordered solid solution) is the main strengthening phase of nickel superalloys, the content of which in modern superalloys reaches up to 0.89. The efficiency of the intermetallic phase as a heat-resistant component in large part limited by low ductility and by strength in a wide temperature range accordingly. An increase in strength limit of the intermetallic component is possible with a decrease in grain size less than a critical value, less than 10 microns. Application of the known methods of plastic deformation for refining an intermetallic grain is almost impossible, but it is physically justified in the period of the grain structure nucleation under non-equilibrium conditions of the exothermic reaction of an intermetallic compound formation in a powder mixture of nickel and aluminum. The retention of low-dimensional grain structure in the synthesized intermetallic compound is possible with combining the processes of crystallization and compaction of the high-temperature synthesis product. This paper presents the results of investigation of the influence of high-temperature synthesis product deformation on the formation of a grain structure in a Ni$_3$Al intermetallic compound synthesized under pressure.

1. Introduction
The conventional technology for producing intermetallic compounds and their alloys is based mainly on melting and casting methods. However, the difficulties of this technology associated with dendritic and zonal segregation in ingots, which determine the relatively low values of strength properties in a cast state led to the development of powder metallurgy technologies. The most significant results were obtained in the development of technologies for obtaining the Ni$_3$Al intermetallic compound with high values of thermal conductivity and the ratio of strength to the specific weight [1], and high resistance to high-temperature oxidation [2]. These technologies include the methods of vacuum sintering under pressure [3], high-temperature impact compaction of a powder intermetallic compact [4], mechanical doping [5], and electric spark plasma sintering [6] of the initial powder materials.

The key parameter determining the strength of the sintered intermetallic is the grain size. It is theoretically shown that for ordered intermetallic compounds, which are characterized by low ductility under tension as a result of fracture along the grain boundaries, there is a critical grain size below which an increase in the crack resistance of the polycrystal occurs (the stress intensity coefficient $K_{Ic}$ increases, the intermetallic compound ductility increases) [7]. Using the example of Ni$_3$Al intermetallic samples obtained from foils with a thickness of 300–500 $\mu$m with a grain size of 1 to 83 $\mu$m, it is shown that refinement of a grain of the $\gamma'$ phase is accompanied by an intensive increase in yield strength and tensile strength — when the grain size decreases from 18 to 1.34 $\mu$m, yield and tensile strengths increase from 280 to 1254 MPa and from 350 to 1757 MPa, respectively [8]. It is noted that the most significant increase in tensile strength with a simultaneous decrease in the plasticity of the intermetallic compound

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is observed when the grain size in the intermetallic compound is less than 10 μm. The above refers to the Ni₃Al intermetallic compound samples obtained either by the method of directional solidification followed by cold rolling, or by the methods of controlled deformation of the cast intermetallic in the form of thin foils for use in the manufacture of microcircuits, microsensors, parts of thermal evaporators, chemical reactors, etc. [9]. To obtain bulk samples of high-strength Ni₃Al intermetallic compound of structural purpose with a low-dimensional grain structure, the issue of technological support remains open.

Solving the problem of forming a low-dimensional grain structure in bulk samples of an intermetallic compound is possible by plastic deformation methods under the conditions of nucleation and crystallization of the intermetallic phase in the exothermic reaction process of an intermetallic compound formation in a compact from a powder mixture of initial elements [10, 11]. The thermophysical conditions of the volumetric reaction of an intermetallic compound formation in the powder mixture of initial elements ensure synchronicity of passing of the phase transformations simultaneously in the entire volume of the powder billet, which makes it possible to consolidate individual structural fragments of the high-temperature synthesis product when the intermetallic phase of a given composition reaches crystallization. The mathematical calculations carried out using the example of the Ni₃Al intermetallic compound showed that the application of pressure to the thermally reactive powder mixture of nickel with aluminum during the thermal explosion mode reduces the grain size in the synthesized intermetallic by an order [12, 13].

This paper presents the research results of the influence of deformation during the high-temperature synthesis process under pressure on the dimension of the grain structure of the synthesized Ni₃Al intermetallic compound.

2. Materials and methods

High-temperature synthesis of the Ni₃Al intermetallic compound under pressure was carried out on experimental bench consist of hydraulic press equipped with a high-frequency generator for heating the steel die-mold prior to self-ignition of the compact from 3Ni + Al powder mixture (nickel particle size was 1 ÷ 3 μm, aluminum - 5 ÷ 10 um). Additionally, the experimental stand equipped with a temperature recorder of heating the powder compact, a digital manometer for recording the pressure in the hydraulic system of the main cylinder of the press and the wake-up timer of the working pressure application in the main cylinder of the press (Fig. 1a). The stoichiometric 3Ni+Al powder mixture was placed in a cylindrical steel die-mold with an internal diameter of 58 mm heated by high-frequency currents. Initiation of the expansion stroke of the hydraulic press and compacting of the high-temperature synthesis product occurred automatically according to the specified program of the technological cycle. In order to further deformation the product of high-temperature synthesis in the working space of the die-mold, the product was compacted with partial extrusion through holes of different diameters in the lower part of the mold (Fig. 1b). In place to change the magnitude of the additional deformation, the synthesis product was extruded through holes of various diameters - 3 and 4 mm.

The grain structure of intermetallic samples was studied by optical metallography (Neophot 32) on lamellar specimens cut from the central part of the synthesized compacts (Fig. 2). Metallographic specimens were prepared by mechanical polishing with a gradual decrease in the size of the diamond abrasive particles of 1 micron. Grain structure was detected by argon ion etching at an accelerating voltage of 0.6 kV. Grain size determined by the method of random secants with averaging over 150 measurements. The phase composition of the samples was investigated by X-ray diffraction analysis using a DRON-7 X-ray diffractometer in CoKα radiation at an accelerating voltage of 40 kV and a current of 40 mA.
3. Results and discussion

In Fig. 3 shows the dependences of the pressure applied to the powder compact of initial elements on the time of the high-temperature synthesis of the Ni$_3$Al intermetallic compound under pressure, including with partial extrusion of the synthesis product.
Presented on Fig. 3 dependencies demonstrate identical character. There is high repeatability of nature of the change in the pressure on the thermoreactive powder mixture throughout the phase transformations during the volumetric exothermic reaction, including with extrusion of the synthesis product. The above means that in each investigated variant, including extrusion of the synthesis product, the reaction of the formation of intermetallic in the powder compact of the initial elements takes place in full (Fig. 4). The latter is confirmed by the results of X-ray diffraction analysis presented in Fig. 4 in the form of diffraction patterns obtained from a reference, synthesized under pressure and synthesized under pressure with extrusion of the high-temperature synthesis product of the Ni$_3$Al intermetallic compound.

**Figure 4.** Diffraction patterns from the reference (1), synthesized under pressure (2) and from the synthesized under pressure with partial extrusion of the product of high-temperature synthesis (3) of the Ni$_3$Al intermetallic compound

The main difference of presented dependencies of the pressure value on the time of the synthesis is the increase in the residence time of the thermoreactive system in the melt state (with the minimum pressure value in the die-mold) at an increase in the delay time of applying the compaction pressure to the synthesis product. Herein the increase in the delay time has a significant effect on the final grain size in the intermetallic compound specimens synthesized under pressure. In Fig. 5 shows the dependences of the grain size synthesized under pressure samples of the Ni$_3$Al intermetallic compound including with the extrusion of the synthesis product on the time delay of applying pressure to the high-temperature synthesis product. We can state the following main features of the presented dependencies:
- the initial increasing in the delay time up to 0.3 s leads to an increase in the grain size in the synthesized intermetallic samples,
- regardless of the presence or absence of extrusion of the synthesis product, with a further increase in the delay time,
- the largest grain size is formed in intermetallic samples synthesized under pressure without extrusion of the synthesis product, a decrease in the grain size in the synthesized intermetallic compound occurs
- deformation of the high-temperature synthesis product in the die-mold with its partial extrusion leads to a fold decrease in the grain size in the intermetallic samples synthesized under pressure,
- an increase in the deformation of the high-temperature synthesis product with an increase in the proportion of extruded synthesis product through an opening in the die-mold of increased diameter further reduces the grain size.

In the intermetallic synthesized under pressure: an increase in the diameter of the holes in the mold from 3 to 4 mm leads to a decrease in the grain size in the synthesized intermetallic, in comparison with the samples synthesized under pressure without extrusion, ~ 1.8 times and ~ 2.5 times, respectively.
Figure 5. Dependences of grain size in synthesized under pressure and synthesized under pressure with partial extrusion of synthesis product samples of intermetallic compound Ni$_3$Al on the delay time of compaction pressure application to the product of high-temperature synthesis.

4. Conclusion
The presented results make it possible to substantiate the promising application of intense plastic deformation to form a low-dimensional grain structure in the intermetallic compound Ni$_3$Al under the conditions of combining the exothermic reaction of the intermetallic compound formation in the powder mixture of the initial elements with the deformation of the high-temperature synthesis product. Deformation processes play a key role in the formation of a low-dimensional grain structure and so that a greater extent, that the higher the degree of the synthesis product deformation. It is necessary to conduct comprehensive studies of the intermetallic compound structural-phase states at all stages of the high-temperature synthesis process under pressure with deformation of the synthesis product to detail the patterns of a low-dimensional grain structure formation.

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