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MODELING OF STRUCTURE FORMATION PROCESS IN INTERMETALLIC NiAl ALLOYS DURING THERMOCHEMICAL PRESSING

Mathematical model aimed at obtaining NiAl alloys with a given structure and properties is proposed and implemented, based on the use of data on the features of the physical modeling of the thermochemical pressing process. For a mathematical description of the process of extrusion of a high-temperature synthesis product, it is necessary to determine a system of equations that takes into account the distribution of the thermo-kinetic and rheological properties of the synthesis product in a mold and caliber. High-temperature synthesis of intermetallic compound NiAl in a powder mixture of pure elements in the conditions of thermochemical pressing allows to obtain an intermetallic alloy with an average grain size of ~ 40—50 microns.
***Keywords**: mathematical model; thermochemical pressing; intermetallic alloys; structure; grain size.*

Математична модель, спрямована на одержання сплавів NiAl із заданою структурою та властивостями, пропонується та реалізується на основі використання даних про особливості фізичного моделювання процесу термохімічного пресування. Для математичного опису процесу екструзії високотемпературного продукту синтезу необхідно визначити систему рівнянь, яка враховує розподіл термокінетичних та реологічних властивостей продукту синтезу у формі та калібрі. Високотемпературний синтез інтерметалічної сполуки NiAl в порошковій суміші чистих елементів в умовах термохімічного пресування дозволяє отримати інтерметалічний сплав із середнім розміром зерна ~ 40—50 мкм.
***Ключові слова**: математична модель; термохімічне пресування; інтерметалідні сплави; структура; розмір зерна.*

Problem's Formulation

The process of phase and structure forming at Ni-Al alloys retrieving from intermetallic compounds is considered. It is established that the main characteristics of intermetallic alloys greatly depend on phase structure [1].

At full leaching of nickel aluminides the fcc-nickel is formed with period of lattice typical for massive nickel. The activity of received catalyst is changed with slope opposition to nickel concentration in Ni-Al compounds. It is established, the leaching product $NiAl_3$ is 3—15 times more active then Ni_2Al_3 alloys and surface area of $NiAl_3$ is twice more active then Ni_2Al_3 one. Activation energy: $NiAl_3$ (E_a = 63 kJ/mol), Ni_2Al_3 (E_a = 75 kJ/mol), $NiAl$ (E_a = 92 kJ/mol) and Ni_3Al (E_a = 126 kJ/mol) [2—4].

It provides the best combination of high-temperature properties — strength, creep resistance, with room ones - plasticity and fracture toughness. Apparently, plastic deformation can be effective not only for the production of fine-grained semi-finished products but also for controlling the parameters of the plate structure in NiAl alloys. In particular, it can be used for obtaining plate-like microstructures with a small colony size and nanocrystalline interplanar spacing, which are of great interest [4].

Analysis of recent research and publications

On the basis of the results of the above papers [5–14] and the earlier self-study results of the thermochemical pressing process [5, 8, 14], an attempt was made to determine the basic patterns of deformation and structure formation, to determine the ways and methods of controlling the processes of forming the structure and properties of compressed articles under conditions self-propagating high-

temperature synthesis (SHS). In order to solve the problem, the method of mathematical modeling is used, in the implementation of which can be conditionally distinguished the following main stages: the idealization of internal properties of the given process (object) and external influences (construction of the physical model); mathematical formulation of the behavior of a physical model (construction of a mathematical model); choice of method for studying a mathematical model and conducting this research; analysis of the obtained mathematical result.

In the mathematical description of thermochemical pressing it is necessary to take into account the thermokinetic characteristics of the process, the velocity of the reactant and its macroscopic density. Therefore, in addition to the kinetic equations for the formation of the intermetallic structure, the activation energy and chemical transformation, it is necessary to use the rheological equations used in describing the rhodinamic models, which allows us to carry out numerical calculations of the kinetic dependences of the basic parameters of the process of compressing the product of high-temperature synthesis — the temperature of synthesis, the completeness of chemical transformation, macroscopic the density of the product of synthesis, the level of elastic stresses in the product, the velocity of its melting point stycheskoy deformation and grain size finite product.

Formulation of the research objective

The purpose of this work is to investigate the structure formation in intermetallic Ni-Al alloys by using a complex plastic deformation technology under non-stationary temperature conditions. Also in given work, the using of mathematical model, the interrelation of the structured quantities with deformation characteristics is considered. It allows to obtain materials with increased plasticity by an optimal combination of mechanical properties over a wide range of temperatures.

Statement of the main material

The starting material for SHS synthesis of the intermetallic compound NiAl is a powder mixture of nickel with aluminum, placed in the form of a molding in a closed mold. The powder compactor is warmed up to a given temperature and ignites in the mode of thermal explosion when the external pressure is applied, under the action of which the compression deforms. The plastic deformation ceases when the synthesis product is cooled to a temperature Tk, at which it loses ductility.

For a mathematical description of the process of extrusion of a high-temperature synthesis product, it is necessary to determine a system of equations that takes into account the distribution of the thermo-kinetic and rheological properties of the synthesis product in a mold and caliber. Assuming that the extrusion occurs in conditions of one-sided compression of the synthesis product in the absence of friction on the walls of the mold, we can write the initial equations:

1. Equation of continuity [10]:

$$\frac{\partial(\rho\rho_1)}{\partial t}+\frac{\partial(\rho\rho_1 V)}{\partial z}=0. \tag{1}$$

2. Equation of motion [10]:

$$\rho\rho_1\left(\frac{\partial V}{\partial t}+V\frac{\partial V}{\partial z}\right)=\frac{\partial\sigma_{zz}}{\partial z} \tag{2}$$

with rheological correlations

$$\sigma_{zz}=\left(\frac{4}{3}\mu+\xi\right)\frac{\partial V}{\partial z}, \tag{3}$$

$$\sigma_{rr}=\sigma_{\theta\theta}=\left(-\frac{2}{3}\mu+\xi\right)\frac{\partial V}{\partial z}. \tag{4}$$

where ρ — relative density, ρ_1 — density of the condensed phase, t — time, V — viscous flow velocity, $\sigma_{rr}, \sigma_{\theta\theta}, \sigma_{zz}$ — radial, tangential and axial stresses, μ, ξ — shear and bulk viscosity.

It is assumed that the distribution of relative density in the initial powder mixture is uniform:

$$\rho(z,0)=\rho_0.$$

In the case where the density of the product SHS differs from the density of the original powder mixture

$$\rho_1 = \frac{\rho_c \rho_f}{\alpha\rho_c + (1-\alpha)\rho_f},$$

where ρ_c — initial density of the mixture:

$$\rho_c = \frac{\rho_{Ni}\rho_{Al}}{\rho_{Ni}(1-c_0) + \rho_{Al}c_0},$$

where ρ_{Al} — aluminum density; ρ_{Ni} — nickel density; ρ_f — the density of the reaction product; α — depth of chemical transformation of the intermetallic compound during the synthesis; c_0 — relative mass concentration of titanium in the initial binary powder mixture.

The depth of chemical transformation is defined as

$$\frac{d\alpha}{dt} = f(\alpha)k_o \exp\left(-\frac{E}{RT}\right), \tag{5}$$

where $f(\alpha)$ — kinetic law; k_0 — pre-exponential factor; E — activation energy of a chemical reaction.

To determine the conversion depth (α), the Johnson-Mel-Avrami-Kolmogorov model was used to estimate the kinetics of the formation of new phases and structural components. This model assumes that the appearance of a new phase occurs uniformly throughout the volume, the rate of appearance of a new phase does not depend on its already available quantity [6, 7]. The equation is written in the form:

$$\alpha(t) = 1 - \exp\left(-Kt^n\right),$$

where K — is determined by the rate of growth of the phase in the volume and depends on the temperature and properties of the particular substance, n — parameter determined by the growth pattern of crystallites.

Different values of n correspond to different conditions for the formation and growth of embryos. If the cores are pre-formed and, therefore, they are all present from the very beginning, the transformation occurs only because of the 3-dimensional growth of the nuclei, then n is 3.

The parameter of crystallite growth rate K can be represented in the form:

$$K(T) \sim \exp(-E_a / RT),$$

since the crystallization process is thermally activated.

3. Equation of thermal conductivity [11]:

$$c_1\rho\rho_1\left[\frac{\partial(\rho T_i)}{\partial t} + \frac{\partial(\rho V T_i)}{\partial z}\right] = \frac{\partial}{\partial z}\left[\lambda(p)\frac{\partial T_i}{\partial z}\right] + \rho\rho_1 Q\frac{\partial\alpha}{\partial t} - \frac{2\chi_i}{r_i}(T_i - T_0), \tag{6}$$

where T_i — the material temperature in the matrix ($i=1$) and in the caliber ($i=2$); $\lambda(\rho)$ — dependence on the density of the heat conductivity of the material; χ_i — effective heat transfer coefficient, r_i — radius of the cross section of the matrix and the caliber; $c_1 = (1-\alpha)c_s + \alpha c_{NiAl}$ — heat capacity of the condensed phase; $c_s = c_{Ni}c_0 + c_{Al}(1-c_0)$ — heat capacity of the initial mixture; Q — thermal effect of intermetallide formation reaction Ni-Al; T_0 — initial temperature.

The following assumptions were made to describe the process of compression of an intermetallic synthesis product in the mode of thermal explosion:

– the heating and cooling of the reacting powder system in the working space of the mold proceeds without a temperature gradient;

– the synthesis product is deformed in a homogeneous-stressed state;

– the heat sink from the mold can be neglected;

– the voltage at the upper boundary of the original powder compression in absolute value is equal to the compression force.

Thus, using the equation (1) and the relation (2), (3), the change in the density of the reactive in the mold of the powder system can be written as [10]:

$$\frac{\partial(\rho\rho_1)}{\partial t} = \frac{\rho\rho_1 N}{4/3\,\mu + \xi} \tag{7}$$

where N — the value of the applied pressure.

A equation of the thermal balance of a synthesis product, taking into account a number of assumptions, can take this form [11]:

$$c_1\rho\rho_1 \frac{\partial T}{\partial t} = \rho\rho_1 Q_{NiAl} \frac{\partial \alpha}{\partial t} - \chi_1 \frac{S}{V}(T - T_0), \tag{8}$$

where S — total area of the inner surface of the mold, V — volume of molds.

The equations (7) and (8) allow us to quantify the parameters of the process of thermochemical pressing of the intermetallic compound synthesized under pressure. The process of forming a structure in a synthesized product under pressure is considered in the assumption that the initial grain size corresponds to the size of the initial particles of the refractory component (nickel), i.e. $D_0 = D_{Ni}$ (D_{Ni} — the diameter of the nickel particle).

The kinetics of grain growth as a result of the recrystallization of the synthesized intermetallic product is estimated from the equation [11]:

$$\frac{\partial D}{\partial t} = \frac{K(T_1)}{D^h}, \tag{9}$$

where D — the initial size (diameter) of the grain; $K = k_0 \exp(-E_a/RT)$ — depends on the constant temperature; k_0 — pre-exponential factor; E_a — energy of activation of grain growth; h — degree of magnitude close to 1.

The amount of deformation of the synthesized product during extrusion is determined from the equation:

$$\varepsilon = \frac{r_1^2 - r_2^2}{r_1^2}. \tag{10}$$

Dependence of the grain size of the synthesized product on the degree of its deformation during extrusion is described by the empirical relations [11]:

$$D_\varepsilon = \frac{D}{\sqrt[3]{A\left(\frac{\varepsilon}{\varepsilon_{кр}}\right)^2}}, \tag{11}$$

where $\varepsilon_{кр}$ — the degree of deformation at which the formation of the nucleation of recrystallization occurs ($\varepsilon_{кр} \approx 0{,}1$); A — coefficient of form of intersection of the initial grain ($4\pi/3 < A < 6$).

A quantitative estimate of the grain size in the intermetallic product of the synthesis after extrusion can be carried out using the heat balance equation (7) and having carried out the derivative in time (8) in the derivative in the temperature.

Thus, after carrying out the necessary transformations, for the final grain size of the intermetallic product under the SHS- pressing, we can write:

$$D_k = \sqrt{D_\varepsilon^2 + \frac{c\rho_0\rho_c r_2 R T_{ad}^2}{\chi_2 E_a (T_{ad} - T_0)} k_0 \exp\left(-\frac{E}{RT_{ad}}\right)}. \tag{12}$$

From equation (12) it is evident that the final grain size in the thermochemical pressing product depends on the size of the grain of the product synthesized in the mold, the degree of deformation of the synthesized product during extrusion through the caliber, the adiabatic temperature of the synthesis of the extruded product and the speed of its cooling (depending on the temperature of the press shape, radius of its cross-section and coefficient of heat transfer of the synthesized product with the walls of the mold).

Computer simulation of the hot-deformation processes of intermetallic NiAl alloys is made using the software package Deform. The Deform program is a powerful system for modeling technological processes designed to analyze the three-dimensional behavior of the metal under various pressure processing processes. The program is based on the finite element method, one of the most well-known, reliable and currently used calculation methods. An automatic grid generator allows you to build an optimized finite element grid, thickening it in the most critical areas. In addition, the program provides important information on material flow and temperature distribution during the deformation process, which allows modeling a complete list of pressure processing processes and solving deformation and heat transfer problems. In solving the thermal deformation problem of compressing NiAl alloys into the Deform program, the following output data were integrated: H_0 = 50 mm, r_1 = 25 mm, r_2 = 15 mm, $T_{a\partial}$ *(NiAl)* = 1911 K, T_0 = 300 K, ρ_0 = 0,6, ρ_{Ni} = 8907 kg/m^3, ρ_{Al} = 2700 kg/m^3, μ = 0,14, ρ_{NiAl} = 5870 kg/m^3, c_{NiAl} = 600 J/kg·K, E_a (*NiAl*) = 92,048 kJ/mol, D_{Ni} = 100 μm.

In work [8], based on experimental research's methods of kinetic interaction in intermetallic alloys in SHS conditions, it was established that for obtaining NiAl alloy the activation energy was nearly 79 kJ/mole and pre-exponential coefficient $k_0 = 7.2 \cdot 10^8$ s^{-1}.

In solving the thermoformation problem of compressing NiAl alloys into the Deform program, the following data were integrated:

– the rheological properties of the NiAl alloys σ = f (ε, u, T), obtained experimentally on the Gleeble-3800 complex [6], which makes it possible to carry out numerical calculations of the kinetic dependences of the basic parameters of the process of compression of the product of high-temperature synthesis — the temperature of the system, the completeness of the chemical transformation, the macroscopic density of the synthesis product, the level of elastic stresses in the product, the speed of its plastic deformation and the grain size of the final product.

– parameters of the hydraulic press, according to the passport and the layout of the equipment;

– deformation and velocity (degree of deformation, displacement of the punch, etc.);

– temperature and temporal (thermophysical characteristics of the deformable and material of the technological instrument, coefficients of heat transfer, radiation, duration of pause, etc.).

To simulate the compression of the NiAl alloy, the original finite element grid consisted of 100 elements grouped in a rectangle of 10 elements on one side. The sample in question was a cylinder 60 mm in diameter and 90 mm high.

The simulation results of the stress-strain state of NiAl alloys are presented in Fig. 1. The process of extrusion is characterized by a stress of comprehensive compression, which provides the material the best in these conditions plastic properties. Under the influence of compressive stresses, the material flows in the direction of the largest gradient of stresses — from the surface of the punch, where they have the maximum value, to the caliber of the matrix (Fig. 1), where the normal stresses on the free surface of the tangent material are zero.

Comprehensive uneven compression provides the material with the highest ductility compared to other processes of metal treatment, but this feature of the process is manifested in extremely uneven deformations. In this case, only the compressive voltages acting continuously in the direction of extrusion from the maximum values to zero are not always in full volume of the deformed material. The presence of the difference between the intersections of the container and the caliber of the matrix, the forces of contact friction and other factors leads to the fact that the particles of the material begin to move not only in the directions of the greatest deformation, but also in transverse directions. The latter contributes to the emergence of local (additional) stresses, the magnitude of different, direction and sign, and the emergence of tensile stresses. This is facilitated by the movement of material particles along trajectories of different lengths with velocity, change in the process of passage through different zones. The results of modeling the stress-strain state are shown in Fig. 2.

In the conditions of the synchronization of thermal processes of the SHS and the dynamic compactation of the synthesis product, it is possible to obtain a compact intermetallic alloy with a highly dispersed structure, the size of which is much smaller than that of the alloys obtained by the methods of casting, sintering or shock-wave action on the synthesized product. Grinding of grain of intermetallic alloy in the process of its synthesis under pressure occurs as a result of plastic deformation of the product of synthesis and high cooling rates (Fig. 3). High-temperature synthesis of the in-

termetallic compound NiAl in a powder mixture of pure elements in the conditions of thermochemical pressing at a thermal explosion at a minimum external pressure on the mixture allows obtaining an intermetallic synthesis product with an average grain size of ~ 40—50 μm. An increase in the degree of plastic deformation of the intermetallic product synthesized under pressure in the conditions of the thermochemical pressing allows to reduce the size of the grain in the final product by an order of magnitude and even to form a sub-microcrystalline granular structure in the intermetallic alloy [14]. The graphical interpretation of calculated results is shown on Fig.3.

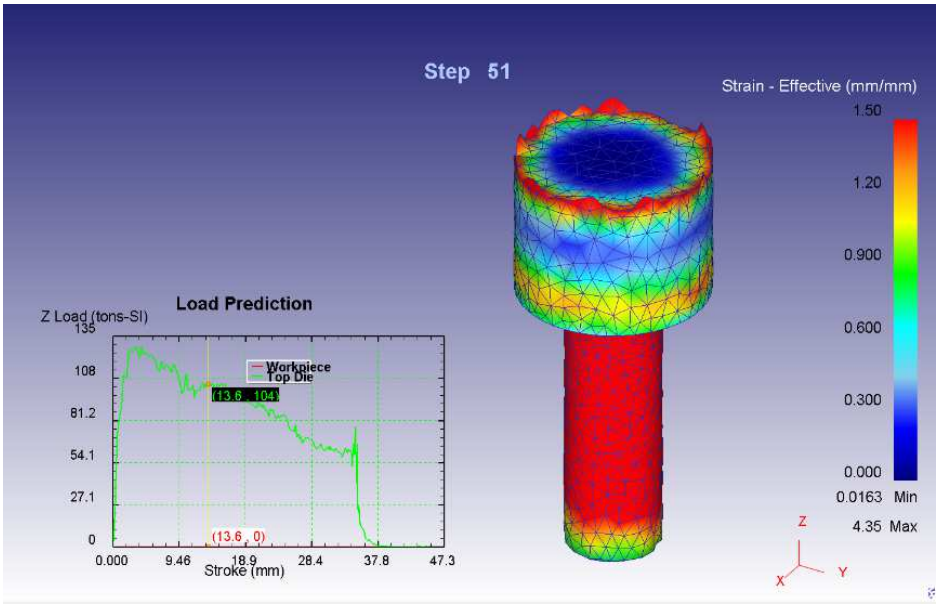

Fig. 1. Simulation of the process of thermochemical pressing of the intermetallic NiAl alloy in the program Deform

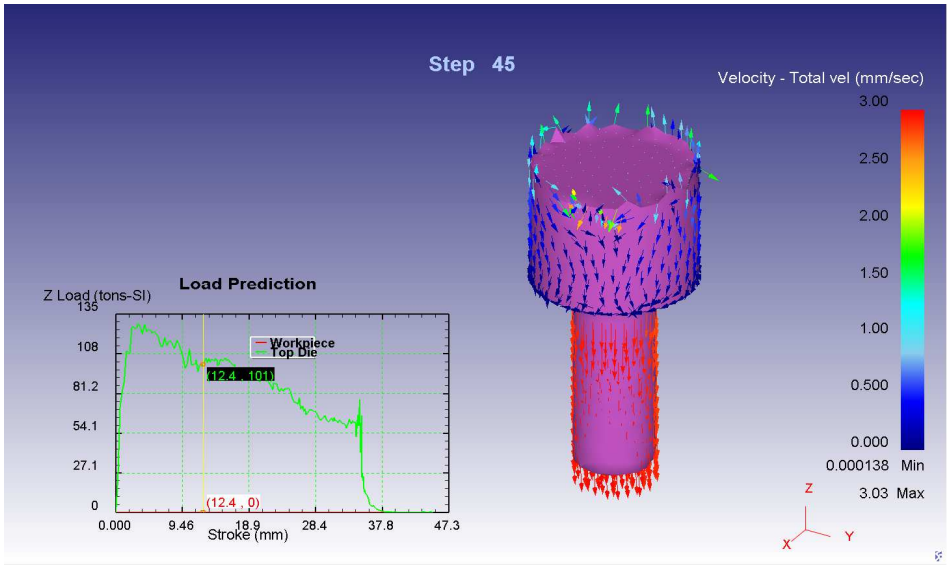

Fig. 2. Modeling of pressing processes of NiAl alloys

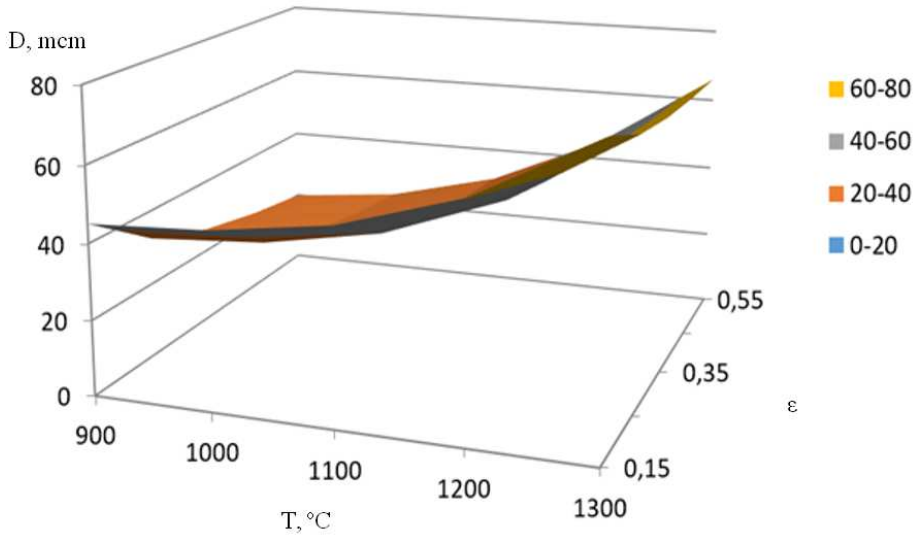

Fig. 3. The dependence of grain size of NiAl intermetallide on deformation and temperature degree

Conclusions

Thus, a mathematical model aimed at obtaining NiAl alloys with a given structure and properties is proposed and implemented, based on the use of data on the features of the physical modeling of the thermochemical pressing process. High-temperature synthesis of intermetallic compound NiAl in a powder mixture of pure elements in the conditions of thermochemical pressing allows to obtain an intermetallic alloy with an average grain size of ~ 40—50 microns.

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МОДЕЛЮВАННЯ ПРОЦЕСУ ФОРМУВАННЯ СТРУКТУРИ В ІНТЕРМЕТАЛІДНИХ NiAl СПЛАВАХ ПРИ ТЕРМОХІМІЧНОМУ ПРЕСУВАННІ

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Реферат

Розглянуто процес формування фази та структури в сплавах Ni-Al, отриманих з інтерметалічних сполуках. Встановлено, що основні характеристики інтерметалічних сплавів сильно залежать від фазової структури. При повному вилуговуванні алюмінідів нікелю утворюється гцк нікель з періодом решітки, характерним для масивного нікелю. Активність отриманого каталізатора змінюється при протилежному нахилі концентрації нікелю в сполуках Ni-Al. Встановлено, що продукт вилуговування $NiAl_3$ в 3—15 разів активніший, ніж сплави Ni_2Al_3, а площа поверхні $NiAl_3$ вдвічі активніша, ніж Ni_2Al_3. Енергія активації: $NiAl_3$ (E_a = 63 кДж/моль), Ni_2Al_3 (E_a = 75 кДж/моль), NiAl (E_a = 92 кДж/моль) і Ni_3Al (E_a = 126 кДж/моль).

Мета даної роботи - дослідити структуроутворення в інтерметалічних Ni-Al сплавах за допомогою складної технології пластичної деформації в умовах нестаціонарних температур. Також в даній роботі, використовуючи математичну модель, розглядається взаємозв'язок структурних величин із характеристиками деформації. Це дозволяє отримувати матеріали з підвищеною пластичністю шляхом оптимального поєднання механічних властивостей у широкому діапазоні температур.

В умовах синхронізації теплових процесів СВС та динамічного ущільнення продукту синтезу можна отримати компактний інтерметалічний сплав із сильно дисперсною структурою, розмір якого значно менший, ніж у сплавів, отриманих методи лиття, спікання або ударно-хвильової дії на синтезований продукт. Подрібнення зерна інтерметалічного сплаву в процесі його синтезу під тиском відбувається в результаті пластичної деформації продукту синтезу та високих швидкостей охолодження. Високотемпературний синтез інтерметалічної сполуки NiAl у порошковій суміші чистих елементів в умовах термохімічного пресування при термічному

вибуху при мінімальному зовнішньому тиску на суміш дозволяє отримати інтерметалічний продукт синтезу із середнім розміром зерна ~ 40- 50 мкм. Підвищення ступеня пластичної деформації інтерметалічного продукту, синтезованого під тиском в умовах термохімічного пресування, дозволяє на порядок зменшити розмір зерна в кінцевому продукті і навіть сформувати субмікрокристалічну зернисту структуру в інтерметалічному сплаві.

Література

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