Impact of High-Temperature, High-Pressure Synthesis Conditions on the Formation of the Grain Structure and Strength Properties of Intermetallic Ni_3Al

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Abstract. The impact of the preliminary load on 3Ni+Al powder mixture and the impact of the duration of the delay in application of compacting pressure to synthesis product under the conditions of continuous heating of the mixture up to its self-ignition on the grain size and strength properties of the synthesized Ni_3Al intermetallide material have been studied. The grain structure of the intermetallide synthesized under pressure was studied by means of metallography, transmission electron microscopy and EBSD analysis, with the dependence of ultimate tensile strength on the grain size in the synthesized intermetallide having been investigated at room temperature and at temperatures up to 1000°C. It is shown that an increase in the pressure preliminarily applied to the initial mixture compact results in reduced grain size of the final intermetallide, whereas an increase in pre-compaction time makes the grain size increased. A decrease in the grain size increases the ultimate tensile strength of the intermetallide. The maximum value of the ultimate tensile strength in the observed anomalous temperature dependence of this strength exhibits a shift by 200°C toward higher temperatures, and the ultimate strength of the synthesized intermetallide at 1000°C increases roughly two-fold.

1. Introduction

Intermetallic compound Ni_3Al provides the basis for a broad spectrum of alloys with improved properties intended for use under thermal and force loads as forging dies, turbocompressor and internal-combustion-engine components, turbine blades in rocket engines, combustion-chamber components, heat exchangers, microreactors, etc. [1]. Enhancement of the application efficiency of intermetallic compound Ni_3Al as the main component of heat-resisting alloys is restricted because of the high susceptibility of the material to brittle fracture; indeed, polycrystalline Ni_3Al suffers brittle rupture along grain boundaries both at low and elevated temperatures [2-5]. Increasing the plasticity and the strength of intermetallic compound Ni_3Al is possible either via doping of the material with various impurities such as boron [6] or via grain-structure refinement [7]. Practical implementation of boron-doping process for intermetallides by traditional methods encounters certain difficulties [8], and enhancement of the mechanical strength of polycrystalline intermetallide samples via grain-structure refinement achieved using severe plastic deformations realized at temperatures as high as the melting point also seem to be impossible because of the high brittleness of the intermetallide at equally high temperatures.

An alternative method for obtaining intermetallic Ni_3Al and alloys based on this intermetallide is the self-propagating high-temperature synthesis (SHS) of the intermetallic compound in a powder mixture of initial elements taken in desired proportion [8-12]. Thermophysical conditions of bulk-uniform synthesis reaction of the intermetallide in the initial powder mixture of pure elements guarantee synchronous proceeding of phase transformations throughout the entire volume of the powder sample. A unique feature of SHS process is the possibility to consolidate individual structural fragments within the synthesis product at the moment the thermo-reacting mixture reaches a homogeneous structural-phase composition at the stage of material crystallization and grain-structure formation [13].

The high rates of phase transformations during the bulk-uniform synthesis reaction in a powder mixture of initial elements yielding the intermetallide entail considerable difficulties in controlling the formation process of the structural-phase state of the synthesis product. The main drawback of known solutions [14,15] hampering the mentioned control is the initiation of high-temperature synthesis of the intermetallide in the initial powder mixture in frontal burning mode, followed by subsequent application of pressure to the synthesis product after the burning front has already traversed the entire volume of the mixture. As a result, during the compaction the synthesis product at its different sections occurs in different structural-phase states, and this circumstance does not allow one to obtain a grain structure with desired grain sizes in synthesized intermetallide. A solution to this problem can only be gained via implementation of synchronization of the bulk-uniform exothermal-reaction processes of intermetallide formation in the powder mixture of initial elements with the dynamic compaction of the synthesis product.

In the present publication, we report on the results of a study of the impact of synthesis conditions on the structural-phase state and strength properties of the intermetallic compound Ni_3Al synthesized under pressure from an initial 3Ni+Al powder mixture whose volume was continuously heated until a self-ignition followed by subsequent dynamic compaction of the synthesis product was reached.

2. Material and methods

The initial materials used for the high-temperature synthesis of intermetallic Ni_3Al samples were mixtures of nickel and aluminum powders in which particle sizes ranged in the intervals 1…3 and 5…10 μm, respectively. The high-temperature synthesis of intermetallic Ni_3Al samples in stoichiometric powder mixture was conducted under a pressure implemented using an automated hydraulic press. The hydraulic press was additionally equipped with a facility for rf heating of the steel mould, with an indicator showing the current heating temperature of the powder compact, with a digital manometer that was used for monitoring the pressure in the main-press-cylinder hydrosystem, and with a timer serving the function of working-stroke initiation (Fig. 1).

The phase composition of synthesized intermetallide samples was studied by means of the X-ray diffraction implemented on a DRON-7 X-ray diffractometer using Co K_α radiation at an accelerating voltage of 35 kV and an electron-beam current of 20 mA. The grain structure of the material was investigated on a Neophot 32 microscope using intermetallic samples sized 40×15×5 mm (these samples were cut out from central parts of the pressure-synthesized pellets). The metallographic sections of the samples were prepared by mechanical grinding performed with gradual reduction of diamond abrasive size down to 1 μm. The grain structure in the samples was revealed by etching the samples with argon ions at an accelerating voltage of 0.6 kV. For evaluation of grain sizes, the random secant technique with the averaging performed over 150 measurements was used.

The TEM study of the material structure was carried out on a JEM-2100 electron microscope at an accelerating voltage of 200 kV. Thin foils were prepared using the ion thinning process implemented on an EM-09100IS (JEOL) facility at an accelerating voltage of 8 kV.

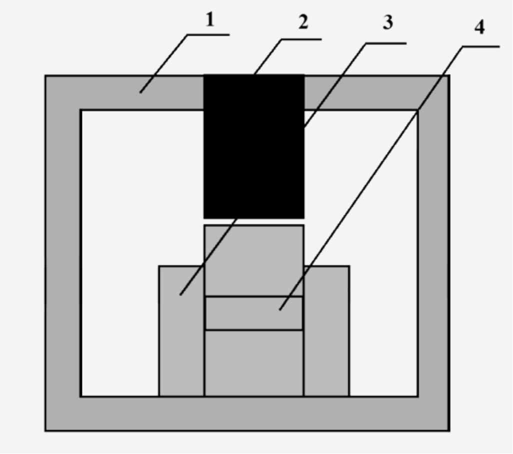

Fig. 1. Schematic of the experimental facility for high-temperature, high-pressure synthesis of chemical compounds and alloys based on such compounds to be conducted in thermal explosion mode during heating of the initial powder compact in a steel mould: 1- press volume, 2 – main press cylinder, 3 – steel mould, 4 – powder compact.

The EBSD analysis of the material structure of the samples was performed on a Quanta 200 3D tungsten-hot-cathode microscope operated at an accelerating voltage of 30 kV. The step size was 0.2 μm. Poor indexed points with the confidence index (CI) values less than 0.1 were discarded from consideration. The cross-sectional cuts of the samples under examination were polished electrolytically at 10°C temperature and 80-V voltage in an electrolyte of the following composition: CH_3COOH – 78%, $HClO_4$ – 22%.

The tensile tests of the intermetallic samples at temperatures 20…1000 °C were performed on LFM-125 walter + baiag testing machines, Switzerland. The deformation rate during the tests was 3×10^{-4} s^{-1}, the dimensions of the gage parts of the test samples being 10×3×1.2 mm.

3. Results and Discussion

The key parameters of the synthesis process of intermetallic compound Ni_3Al held under pressure and proceeding through a bulk-uniform exothermal synthesis reaction are the magnitude of the initial load applied to the powder mixture of nickel and aluminum and the duration of the delay in application of compacting pressure to the synthesis product after ignition of the mixture. Those parameters define the formation kinetics of the grain structure in synthesized products. Programming of the values of both parameters is based on the control of the load being preliminarily applied to the powder compact in the mould exerted using preliminary fixation of a set pressure value in the main-press-cylinder hydrosystem and a set duration of the delay in the initiation of the press working stroke used to compact the synthesis product. Figure 2a shows a diagram illustrating the variation of the pressure applied to the powder compact in the steel mould. This diagram was obtained by recalculating the pressure values in the main-press-cylinder hydrosystem in coordinates "pressure-time" over the entire cycle of the high-temperature synthesis process of the intermetallide in the powder mixture continuously heated in the steel mould. At an initial pressure P_o implemented during continuous heating of the mixture, the plastic deformation of the powder compact, the melting of the aluminum component, and the initiation of the exothermal synthesis reaction all resulted in a reduction of the pressure applied to the powder compact down to a pressure P_1. Further scenario implies either an immediate compaction of the synthesis product under the pressure P_2 implemented due to initiated press working stroke or preliminary storage of the mixture during some time under pressure P_1 with subsequent compaction of the synthesis product. Figure 2b shows the experimental time dependences of the pressure applied to the powder compact being continuously heated in the steel mould until reaching self-ignition with various pre-compaction times. In accordance with the indicated regimes of the high-temperature synthesis of the intermetallide under pressure, intermetallic Ni_3Al samples were obtained at different magnitudes of the preliminary load exerted on the initial powder mixture and at different times during which the synthesized product was held prior to the moment of its compaction.

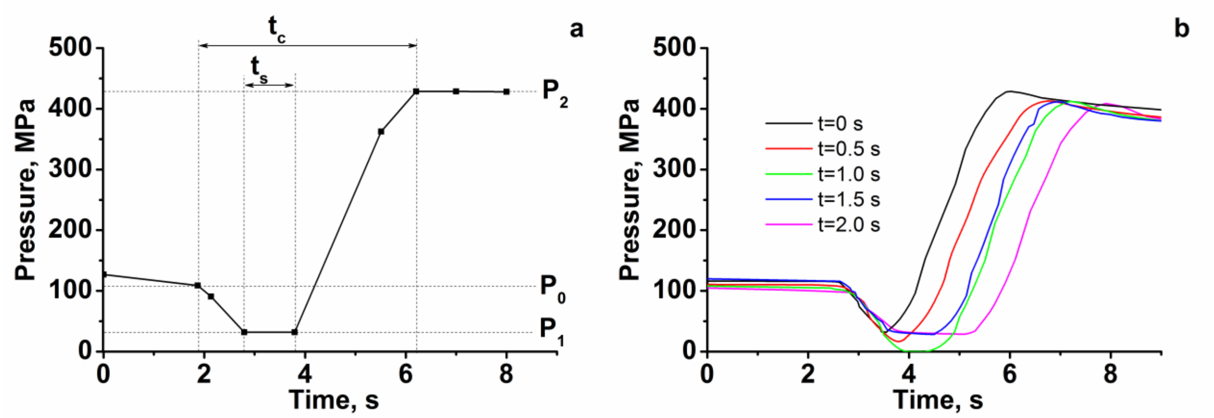

Fig. 2. A diagram of pressures applied to 3Ni+Al powder compact plotted in coordinates “pressure-time” over the entire process cycle of the high-temperature synthesis of Ni_3Al intermetallic compound from an initial powder compact being continuously heated under a preliminary load in the steel mould with a time delay in application of compacting pressure (a) and experimentally obtained diagrams illustrating the variation of pressure during the entire programmed cycle of the synthesis process of Ni_3Al intermetallide under continuous heating of the powder mixture implemented with various pre-compaction times (b).

3.1. Impact of the magnitude of the preliminary load on the phase composition and grain size in the pressure-synthesized Ni_3Al intermetallide

X-ray data obtained for intermetallic Ni_3Al samples synthesized at extreme pressure values in the examined range of initial pressures applied to the powder compact, 33 and 115 MPa, without any delay in time in application of the minimum compacting pressure (diffractograms 1 and 2, respectively) are shown in Fig. 3.

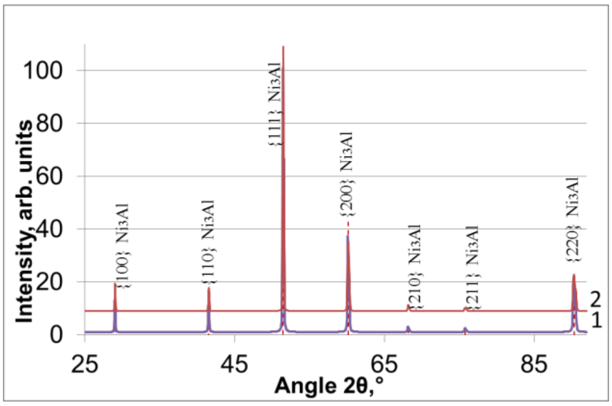

Fig. 3. Diffractograms of intermetallic Ni_3Al samples synthesized under pressure from an initial powder mixture continuously heated in the steel mould at different pressures applied to the 3Ni+Al powder compact, 33 MPa (diffractogram 1) and 115 MPa (diffractogram 2) with zero pre-compaction time.

A comparison between the diffractograms of Fig. 3 shows that the change in the magnitude of the initial pressure applied to the initial powder mixture does not affect the phase composition of the synthesized intermetallic samples, - in both cases, the phase composition corresponds to the intermetallic compound Ni_3Al.

The change in the magnitude of the initial pressure has an impact on the grain size of the pressure-synthesized Ni_3Al intermetallide. Figure 4 shows the general appearance of the grain structure in intermetallic Ni_3Al samples synthesized under pressure at different values of the pressure that was preliminarily applied to the initial powder compact kept in the mould. Numerical estimations of the dependence of the grain sizes in the pressure-synthesized intermetallide samples show that, with increasing the initial pressure, the grain size in synthesized intermetallides diminishes from 8.5 μm down to 1.5 μm (see Fig. 5).

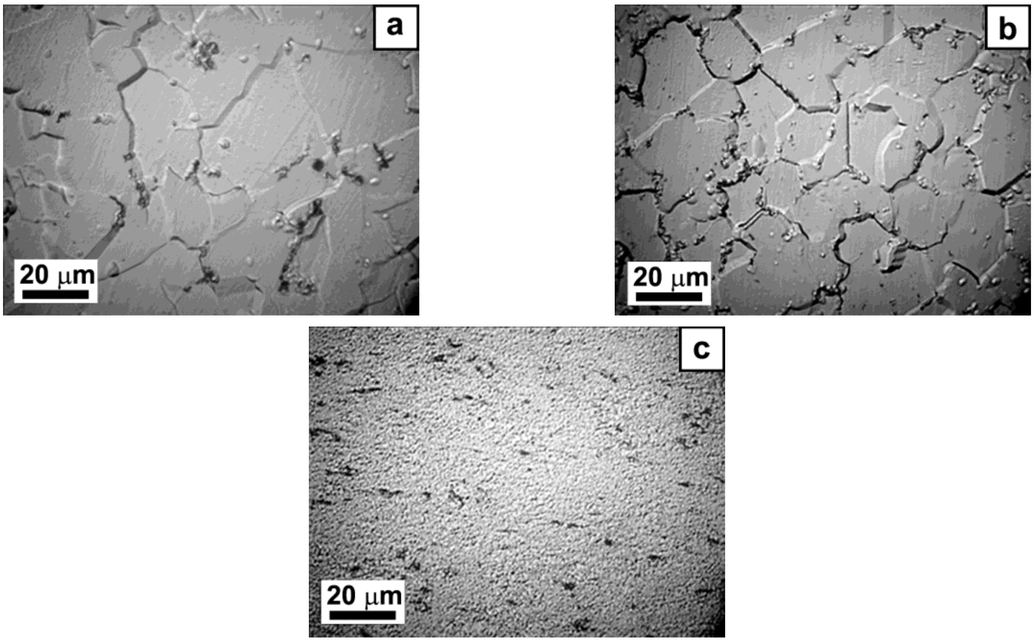

Fig. 4. The grain structure of intermetallic Ni_3Al samples synthesized under pressure at different values of the pressure that was preliminarily applied to the initial powder mixture: a – P_o= 33 MPa, b – P_o= 76 MPa, c – P_o= 115 MPa.

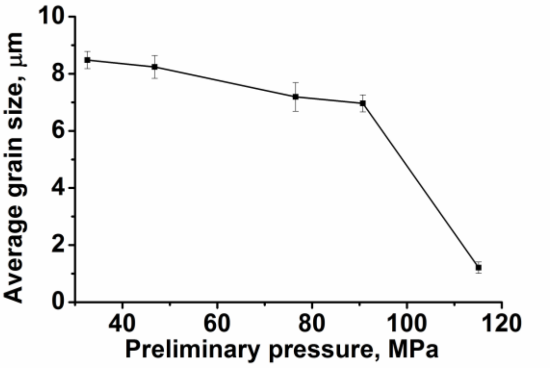

Fig. 5. The average grain size in the pressure-synthesized intermetallic Ni_3Al samples versus the magnitude of the pressure preliminarily applied to the initial powder mixture.

The TEM study of the microstructure of pressure-synthesized Ni_3Al intermetallides and their EBSD analysis have confirmed the data gained while performing the metallographic study of the grain structure, - it was found that the grain sizes in the grain structure formed in the intermetallide under preliminary 115-MPa loading of the initial powder mixture fell into the micrometer range (see Fig. 6).

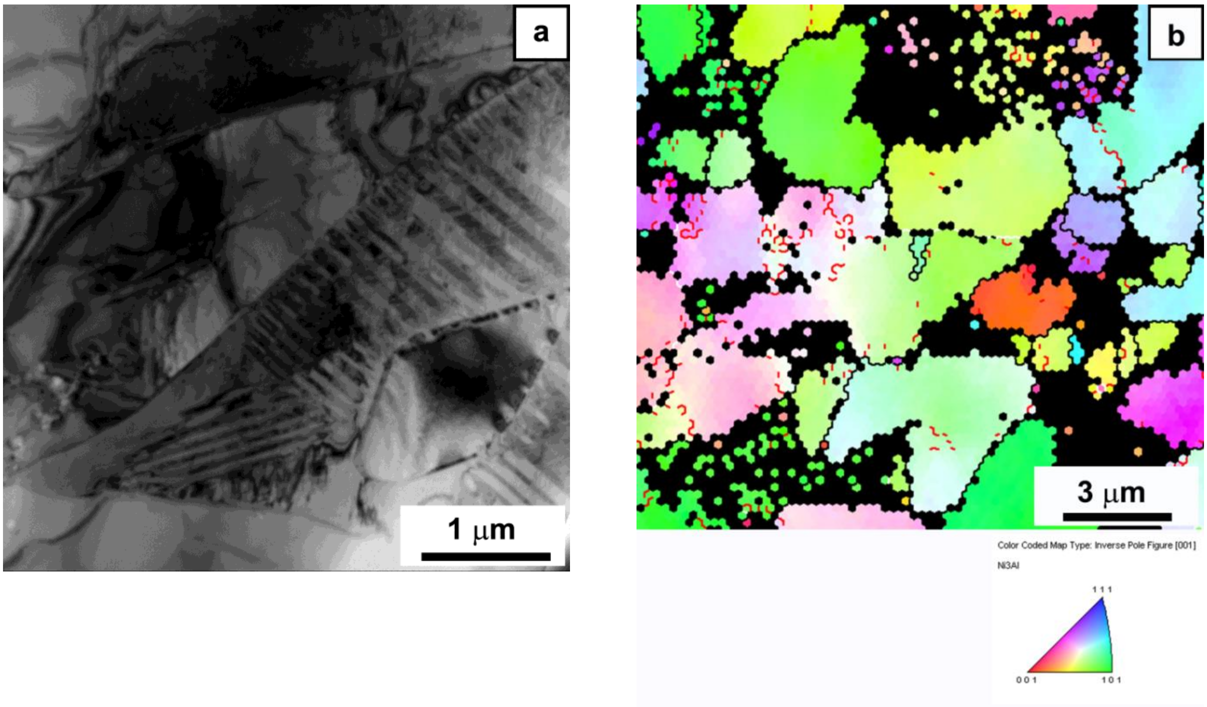

Fig. 6. The grain structure of the intermetallic Ni_3Al synthesized under pressure at 115-MPa pressure preliminarily applied to the initial powder mixture (as revealed by TEM) (a) and the inverse pole figure demonstrating the grain structure of the intermetallide. The black and red lines show the high-angle and low-angle boundaries, respectively (b).

3.2. Impact of pre-compaction time on the phase composition and grain size in pressure-synthesized Ni_3Al intermetallide

Diffractograms of intermetallic Ni_3Al samples synthesized under pressure at different pre-compaction times are shown in Fig. 7.

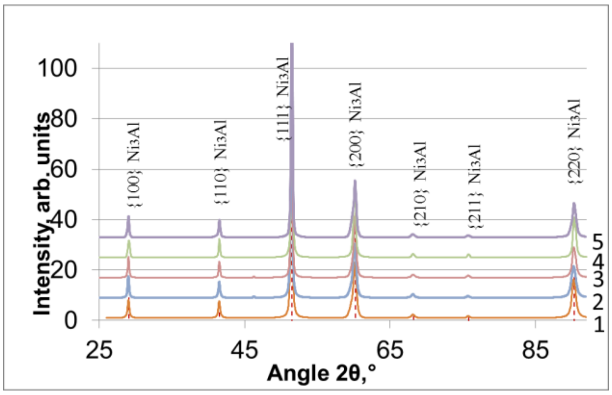

Fig. 7. Diffractograms of intermetallic Ni_3Al samples synthesized under pressure at different pre-compaction times: 1 – 0 s, 2 – 0.5 s, 3 – 1.0 s, 4 – 1.5 s, 5 – 2.0 s.

The increase in pre-compaction time from 0 to 2.0 s has no influence on the phase composition of the synthesized intermetallic material, - indeed, the intermetallides synthesized at all pre-compaction times presented a single-phase Ni_3Al product. However, with increasing the pre-compaction time the average grain size of the intermetallide clearly increases (see Figs. 8 and 9).

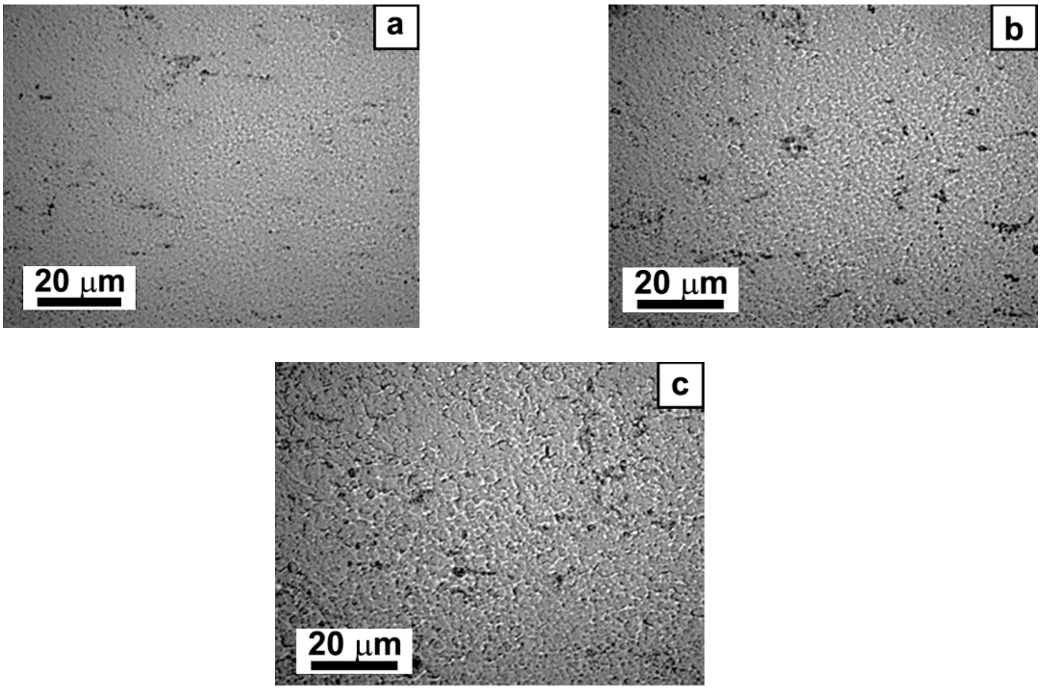

Fig. 8. The grain structure of intermatallic Ni_3Al samples synthesized under pressure at different pre-compaction times: a – 0 s, b – 1 s, c – 2 s.

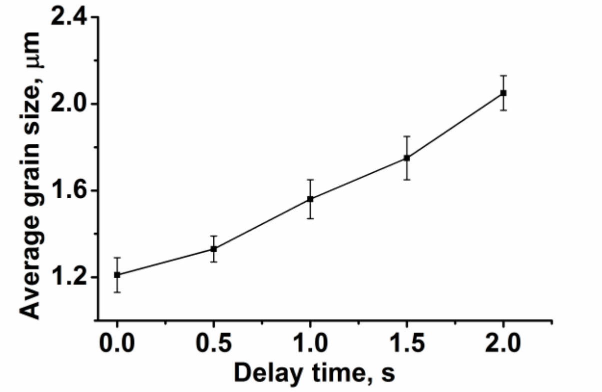

Fig. 9. The grain size in intermetallic Ni_3Al samples synthesized under pressure versus the pre-compaction time.

3.3. Impact of grain sizes on the strength properties of pressure-synthesized Ni_3Al intermetallides

The variation of grain sizes largely affects the strength properties of pressure-synthesized Ni_3Al intermetallides. The ultimate strength of intermetallides under tension versus the grain size in the material and the temperature dependences of the ultimate tensile strengths in the materials with grain sizes 1.2 μm and 8.5 μm are shown in Fig. 10 (a and b, respectively).

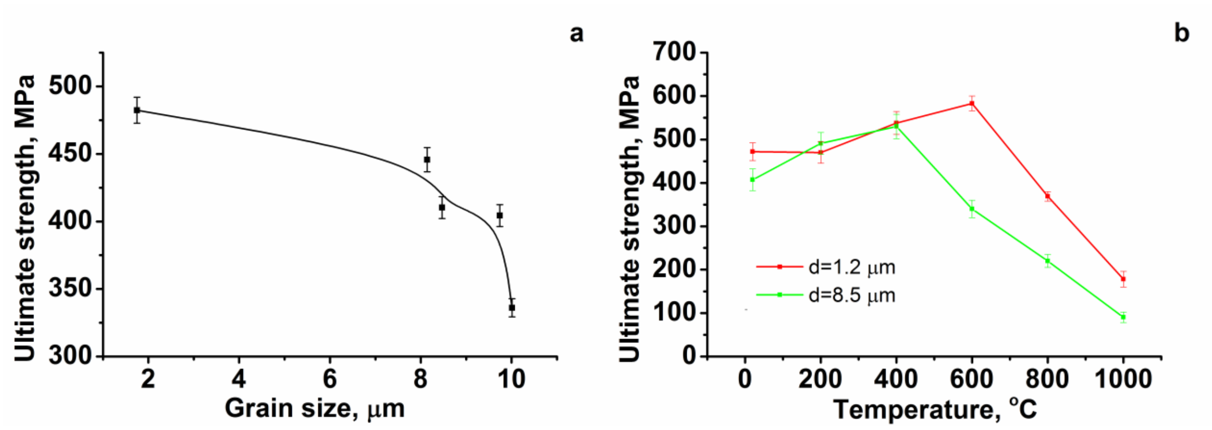

Fig. 10. The ultimate tensile strength of pressure-synthesized Ni_3Al intermetallides versus the grain size (a) and versus the temperature at which the tests were performed (in the materials with grain sizes 8.5 μm (green) and 1.2 μm (red) (b).

The decrease of the grain size in pressure-synthesized Ni_3Al intermetallides from 8.5 down to 1.2 μm increases the ultimate strength of the intermetallide under tension from 336 to 482 MPa (i.e., by a factor of 1.4). In the temperature dependence, the maximum ultimate strength increases from 538 to 583 MPa, and it exhibits a shift by 200 °C toward higher temperatures. At a temperature of 1000 °C, the ultimate strength of the intermetallide under tension increases from 90 to 178 MPa (i.e. roughly two-fold).

4. Conclusion

Programming of main process parameters of the high-temperature, high pressure synthesis of Ni_3Al intermetallide under continuous heating of the initial 3Ni+Al powder mixture prior to reaching its self-ignition provides an efficient means for controlling the formation process of the grain structure in the synthesized intermetallides. Indeed, an increase in pre-compaction pressure decreases the grain size in the synthesized intermetallide. On the contrary, an increase in pre-compaction time increases the grain size in the synthesized product.

A decrease in the grain size increases the room-temperature ultimate strength of the intermetallide under tension. The maximum ultimate tensile strength of the intermetallide in the anomalous temperature dependence of this strength exhibits a shift by 200 °C toward higher temperatures, and the ultimate tensile strength of the intermetallide at a temperature of 1000 °C shows roughly a two-fold increase.

Acknowledgements

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