Stimulation of processes of self-propagating high temperature synthesis in system Ti + Al at low temperatures by influence of γ-quanta

A V Sobachkin¹, M V Loginova¹, A A Sitnikov¹, V I Yakovlev¹, V Yu Filimonov¹ and A V Gradoboev²

¹ Polzunov Altai State Technical University, 46, Lenina Av., Barnaul, 656038, Russia
² National Research Tomsk Polytechnic University, 30, Lenina Av., Tomsk, 634050, Russia

E-mail: anicpt@rambler.ru

Abstract. In the present work, the influence of the irradiation with gamma-quanta ⁶⁰Co upon the structural and phase state of the components of the mechanically activated powder composition of Ti+Al is investigated. The phase composition, structural parameters, and crystallinity are examined by means of X-ray diffractometry. It is found out that the irradiation with gamma-quanta changes the structure of the mechanically activated powder composition. The higher irradiation dose, the higher the structure crystallinity of both components with no change in phase state. At the same time, the parameters of Ti and Al crystal lattices approach to the initial parameters observed before the mechanical activation. The irradiation with gamma-quanta leads to decrease of internal stresses in the mechanically activated powder composition while nanocrystallinity of the structure remains unchanged. Using of powder compositions exposed to the irradiation with gamma-quanta for the SH-synthesis helps to increase speed of the reaction, decrease the peak firing temperature and improve homogeneity, as well as the main phase of the produced material is TiAl.

1. Introduction

Today, along with the traditional methods of material modification (thermal and chemical) giving the possibility to change physical and mechanical properties of the material, increasing attention is paid to extreme methods that lead to forming nonequilibrium nanostructural states in material. The transformation of the material structure to a nanoscale state is able to result in occurrence of unusual physical and mechanical properties that are essentially differ from the properties of coarse-grained materials, and it represents a great practical interest [1-4].

One of the effective ways of the external influence upon materials for the purpose of submicrocrystalline structure creation is a mechanical activation, in which case the great doses of additional energy are introduced into the system that are responsible for its metastable condition. Given that one part of the energy is reserved in crystalline defects and the other part is used to increase surface energy by means of the grain size decrease [5-10].

At the present day, the most effective way of the new composite material obtaining consists in combination of preliminary mechanical activation and subsequent self-propagating high-temperature synthesis (hereinafter referred to as SH-synthesis) [11-13].
An ionizing radiation is also capable to change physical and mechanical properties of the material by means of modification of its structure [14-16]. In this case, the ionizing radiation leads to radiation-stimulated reconstruction of the initial defective structure, taking down the local mechanical stresses with partial annealing of existing defects, and improving the homogeneity of electrophysical and structural properties [12, 17-20].

Moreover, depending on structural condition of the initial materials, the ionizing radiation is able to form a special nonequilibrium state that is notable for grain size decrease and substructural parameters change [21, 22].

In practice a radiation-stimulated diffusion is also observed. It brings the material structure into a more ordered state [23, 24].

It should be pointed out that literature data analysis has brought to light lack of information on the influence of ionizing radiation upon powder composition activation, SH-synthesis parameters and properties of produced materials.

Therefore, the purpose of the present work is the investigation of the influence of gamma-quanta \(^{60}\text{Co}\) irradiation of Ti+Al powder composition with and without the preliminary mechanical activation upon the structural and phase state of the powder composition components and the main parameters of subsequent SH-synthesis.

2. Materials and methods

The powder composition of Ti+Al has been chosen as an object under investigation because titanium aluminides and alloys on their base are used increasingly frequently as structural materials for work in extreme conditions [25-29]. It is possible to use the obtained experimental results as a processing technology for nanocomposite composition modification [30-31].

A simple powder composition of Al (36%wt) + Ti (64%wt) was prepared from titanium powder with the average particle size of 50±10 \(\mu\)m and aluminium powder with the average particle size of 12 \(\mu\)m. Subsequently the mechanical activation during the 7 minutes was conducted in the planetary ball mill under normal acceleration of 40 g. The grinding bodies-to-initial material weight ratio was 20:1. To eliminate oxidation, air from the mill cylinders was pumped out and then the cylinders were filled up with argon under the pressure of 0.3 MPa. After the mechanical activation, the powders were taken out from the cylinders inside a special box, in argon atmosphere [32-33].

Then the mechanically activated powder composition was used to produce cylindrical samples with the diameter of 10 mm and height of 5 mm. The pressing of samples was conducted under room temperature by means of the conventional laboratory press with the load of 40 kN.

The irradiation with gamma-quanta \(^{60}\text{Co}\) was conducted in the certificated fixed system under normal climatic conditions. The level of gamma-quanta radiation was determined with absorbed dose \(D_\gamma\) [Gy].

For structural phase analysis, the general purpose X-Ray diffractometer DRON-6 on the base of copper radiation [CuK\(\alpha\) (\(\lambda=0.15418\) nm)] was used. The diffraction patterns of all samples were registered under the same conditions that gave an option of comparing obtained values in more specific way. Scan step \(h\) during the measurements was equal to 0.05° and the exposure time was equal to 3 seconds. The processing and analysis of the experimental data were conducted by means of PDWin software. For the fine structure parameter determination, the Size&Strain utility of the PDWin software was used, with adjustment for instrumental broadening. Grain sizes and microstrains were determined as the coefficients of the equations set by means of the least absolute deviation method.

The investigation of samples microstructures was conducted on the polished sections using the metallographic electron microscope Carl Zeiss EVO50 XVP.

At the final stage, the experimental SH-synthesis of the Al+Ti powder compositions under investigation was conducted (for initial material, mechanically activated one, and mechanically activated one with subsequent irradiation with gamma-quanta). The synthesis was implemented at the volume firing by means of induction heating that is capable to generate electromagnetic energy in a broad power range. The diagram of the installation used for the SH-synthesis is shown in Figure 1.
In all cases, the high-temperature synthesis was conducted under the same conditions in the following way (see Figure 1). Powder composition 1 was put into black-lead crucible 2. The composition together with thermocouple 4 was consolidated with the laboratory press under the pressure. After that the black-lead crucible with the pressurized powder and the thermocouple was put into induction coil 3 in vacuum chamber 5. Then the SH-synthesis was initiated by means of composition heating with quickly alternating electromagnetic field. For the measuring of the synthesis temperature, the tungsten perrhenic thermocouples connected to the multichannel ADC board combined with PC were used.

3. Results and discussion

3.1 The investigation of the mechanically activated powder compositions

Let us consider the results of the investigation of the mechanically activated powder compositions with and without subsequent irradiation with gamma-quanta. In Figure 2, there are diffractograms of the Ti+Al powder compositions obtained at all stages of the research.

Let us consider the changes at the diffractograms driven by the mechanical activation of the powder composition. After the mechanical activation of the initial powder composition at the diffractogram (Figure 2b), the Ti and Al diffraction reflections of low intensity and increased diffuse background are seen as compared with the diffractogram of the initial composition. That gives evidence of small grain sizes and presence of nonequilibrium defects in the ground material. The additional formations do not appear after the mechanical activation.
As a gamma-quanta irradiation dose becomes higher (Figure 2c, d, e), the increase of the composition components structure crystallinity up to the initial values is seen (at the peak irradiation dose the intensity of diffraction reflections from the irradiated sample is close to the intensity of peaks of the initial composition). However, a little relative peak broadening is preserved that may give evidence of the retention of crystalline nanostructural state as well as the presence of residual microstrains.

The Ti and Al fine structure determination approved that with the change of the irradiation dose, the coherent scattering region (crystalline size) of D titanium and aluminium changes too, as it is shown in Figure 3. It has also been established that with the gamma-quanta irradiation dose growth, the microstrains of mechanically activated composition lower (Figure 4).

The calculation of the precise parameters for the irradiated and mechanically activated samples has shown that after the irradiation with gamma-quanta, the structure crystallinity grows and the structural state of the components changes. Furthermore, under the irradiation, the Ti lattice parameters grow while the Al lattice squeezes.

Comparing the diffraction patterns and calculated data allows assuming that irradiation with gamma-quanta leads to the partial annealing of the defects (Figure 4). However, the grains of both components retain their nanostructural state (Figure 3). From there, the investigation results shown above allow concluding that the ionizing radiation leads to a decrease of local mechanical stresses and annealing of crystalline material structural defects. This matches the investigation results published earlier [14, 18-20].

In Figure 5 the microstructure of the composition samples under investigation is shown. The image is obtained by means of electron microscope. Under the low irradiation dose (Figure 5a), the non-homogeneous structure in the form of eutectics with the high degree of dispersion is observed. Herewith an essential part of light titanium inclusions of prolate form is present. They are homogeneously distributed in the aluminium matrix that has fibrelike structure of dark color. With the irradiation dose growth (Figure 5b), the structure becomes more homogeneous and the quantity of titanium inclusions increases as compared with the small dose area. In both cases, the two different phases have no definite border between them and the general sample structure is vesicular.

The research mentioned above shows that the preliminary mechanically activated powder compositions have non-homogeneous state that is characterized by excess energy induced by
submicron sizes of grain and the presence of microstrains.

Figure 5. The microstructure of mechanically activated powder composition: (a) – $D\gamma = 2\cdot10^3$ Gy; (b) – $D\gamma = 2\cdot10^4$ Gy.

The irradiation with gamma-quanta provides an opportunity to change structural state of the mechanically activated Ti+Al compositions through the modification of precise parameters of the composition components lattices and their fine structure. In particular, the change of irradiation dose makes it possible to stabilize the structure of the mechanically activated composition, decrease stress, perform radiation defects annealing, and retain the nanocrystallinity.

Therefore, the ionizing radiation (in particular, gamma-quanta) gives an opportunity to change intentionally the structural properties of mechanically activated powders, decrease their level of defectiveness, and improve homogeneity of their properties.

3.2 The investigation of SH-synthesis

The next step of the research is a high-temperature synthesis of the Ti+Al powder composition described above.

In all cases, the SH-synthesis was conducted under the same conditions with thermal explosion obtained by means of inductive heating. The heating source was disconnected at the moment when the composition achieved the peak temperature. Then the composition cooled down to the room temperature. The diagram of the installation is shown in Figure 1.

In Figure 6, the SH-synthesis thermograms for corresponding powder compositions are shown. As it is seen, in the case of simple mechanical powder composition (Figure 6a) the heating reaction begins at the aluminium melting point that is equal to about $660^\circ$C.

The preliminary mechanical activation of the powder composition increases its reactivity that leads to the acceleration of chemical reactions (Figure 6b). In this case, the reaction starts at solid phase and the heating reaction starts at the temperature that is close to the temperature of the environment. In the case of the SH-synthesis of the mechanically activated and gamma-quanta irradiated powder composition (Figure 6c), the reaction starts with the aluminium melting as in the case of the initial powder composition that was not subject to any processing (Figure 6a). The solid-phase reaction that is present in the case of the simple mechanically activated composition (Figure 6b) does not take place here. However, as opposed to the first case (Figure 6a), the reaction speed increases and the peak firing temperature decreases ($T_{\text{max}}=1030$ °C in this case, and $T_{\text{max}}=1100$ °C in the case of the simple composition).

In Figure 7, the diffractograms of the SH-synthesis products produced from the different powder compositions are shown. The diffractograms analysis approves that in the cases of the initial powder composition synthesis and the mechanically activated powder composition synthesis, the end products are TiAl, TiAl$_3$, Ti$_3$Al, and there is residual $\beta$-Ti.

The content of TiAl$_3$ in the sample of the mechanically activated composition is lower as compared to the sample of the initial powder composition. In this case, the observed low peak intensity and peak broadening give evidence of non-homogeneous state of the structural components. In the case of
The synthesis of the mechanically activated composition with subsequent irradiation with gamma-quanta, the main phase is TiAl, while the combinations of TiAl$_3$ and Ti$_3$Al are present in small quantities.

Figure 6. The SH-synthesis thermograms for the powder compositions: (a) – the initial composition; (b) – the mechanically activated composition; (c) – the mechanically activated composition at $D_{\gamma} = 1 \cdot 10^3$ Gy

Figure 7. The diffractograms of the products of the Ti+Al powder composition SH-synthesis: (a) – the initial composition; (b) – the mechanically activated composition; (c) – the mechanically activated composition at $D_{\gamma} = 1 \cdot 10^3$ Gy.

Therefore, the irradiation with gamma-quanta of the mechanically activated Ti+Al powder composition gives opportunity to change the parameters of the SH-synthesis. Herewith the portion of the main product TiAl increases, while the secondary phases TiAl$_3$ и Ti$_3$Al decrease, and the homogeneity of the product obtained gets better.

The investigation results shown above allow concluding that the gamma-quanta $^{60}$Co irradiation of the mechanically activated Ti+Al composition is capable to change the SH-synthesis parameters in an appropriate manner in order to obtain the better product.

It is worth to note that such an influence can be expected from the other types of ionizing radiation. Obviously, for use in practice irradiation with gamma-quanta of powder compositions intended for the SH-synthesis, it is important to determine the optimal irradiation dose.

The same influence can be expected from ionizing radiation (in particular gamma-quanta radiation) not only for other powder materials intended for the SH-synthesis, but also in the case of using other powder technologies.

4. Conclusion
Let us summarize the core results and conclusions obtained in this research.

1. The gamma-quanta irradiation of the mechanically activated Ti+Al powder composition changes its physical and mechanical properties through the modification of the composition components structural state.

2. The gamma-quanta influence leads to the improvement of composition components structural states that were violated by the mechanical activation. The evidence for this is the increase of the diffraction reflection intensity of the composition components, and turning back of the lattice parameters to the initial state. Herewith, the Ti lattice parameters grow while the Al lattice parameters...
decrease.

3. The gamma-quanta influence stimulates radiation annealing of nonequilibrium defects in the mechanically activated composition while the grain sizes remain unchanged.

4. The microstructure of the samples produced from the mechanically activated composition with subsequent irradiation with gamma-quanta by means of pressing has no definite borders between composition components due to their redistribution as the result of radiation-stimulated diffusion.

5. The gamma-quanta irradiation of the mechanically activated Ti+Al powder composition changes the parameters of the high-temperature synthesis (the reaction rate increases, the peak firing temperature decreases), and the main phase of the obtained product is TiAl.

5. Acknowledgments

The work was supported by the Ministry of Education and Science of the Russian Federation (Zadanie № 11.1085.2017/4.6).

References

[1] Leyens C and Peters M 2003 Titanium and Titanium Alloys. Fundamentals and Applications (Wienheim: Wiley-VCH Verlag GmbH)
[2] Salishchev G A, Galeev R M, Malyshova S P, Zherebtsov S V, Mironov S Yu, Vafiakhmetov O R and Ivanisenko E V 2006 Metal Science and Heat Treatment 48, 1-2 63-69
[3] Dao M, Lu L, Asaro R J, De Hosson J T M and Ma E 2007 Acta Mater. 55 4041-65
[4] Valiev R Z and Aleksandrov I V 2007 Obemnye nanostructurnye metallicheskie manerialy (Moscow: Akademkniga)
[5] Valiev R Z and Aleksandrov I V 2000 Nanostructurnye materialy, polychennye intensivnoi plasticheskiy deformacii (Moscow: Logos)
[6] Koch C C 1989 Ann. Rev. Mater. Sci. 19 121-143
[7] Boldyrev V V 2006 Russ. Chem. Rev. 75, 3 177-216
[8] Loginova M V, Filimonov V Yu, Yakovlev V I, Sytnikov A A, Negodyaev A Z and Shreifer D V 2015 Applied Mechanics and Materials 788 117-122
[9] Takaki S 2003 Mater. Scie. For. 4 215-222
[10] Dmitriev A I, Zolnikov K P, Psakhie S G, Goldin S V, Lyakhov N Z, Fomin V M and Panin V E 2001 Phys. Mesomech. 4, 6 57-66
[11] Bartolotta P A and Krause D L 1999 Proc. The Second Int. Symp. Gamma Titanium Aluminides 3
[12] Mukasyan A S 2005 Proc. of the Combustion Institute 30 2529-35
[13] Filimonov V Y, Evstigneev V V, Afanas'ev A V and Loginova M V 2008 International Journal of Self-Propagating High-Temperature Synthesis 17, 2 101-105
[14] Gradoboev A V and Surzhikov A P 2005 Radiation stability of GaAs microwave devices (Tomske: Izdatelstvo TPU)
[15] Kyrzina I A, Kozlov E V and Charkeev Yu P 2008 Nanocrystallicheskie intermetallidnye b nitridnye structures (Tomske: Izdatelstvo NTL)
[16] Mamontov A P and Chernov I P 2009 Effect malych doz ioniziryuchego izluchenia (Tomske: Delteplan)
[17] Gradoboev A V and Orlova K N 2015 Phys. Status Solidi C 12, 1-2 35-38
[18] Torkhov N A, Gradoboev A V and Mihalitskyi M M 2012 22nd Int. Crimean Conference Microwave & Telecommunication technology 647-648
[19] Gradoboev A V and Orlova K N 2014 24th International Crimean Conference Microwave & Telecommunication technology 874-875
[20] Gradoboev A V and Orlova K N 2015 IOP Conf. Series: Materials Science and Engineering 81 012008
[21] Lu, Faulkner F G, Jones R B and Flewitt P E J 2005 Journal of ASTM International 2, 8 180-194
[22] Chmelevskaya V S, Bogdanov N Yu and Kordo M N 2008 Physics and Chemistry of Material Treatment 2 14-18
[23] Chalaev A M 1972 Radiachionno-stimylirovannaya diffusia v metallach (Moscow: Atomizdat)
[24] Djafarov T D 1991 Radiachionno- stimylirovannaya diffusia v pluprovodnikah (Moscow: Energjatomizdat)
[25] Il’in A A, Kolachov B A and Pol’kin I S 2009 Titanovye splavi. Spravochnic (Moscow: VILS – MATI)
[26] Germann L, Banerjee D, Guedou J Y and Strudel J-L 2005 Intermetallics 13 920-924
[27] Sobachkin A V, Sitnikov A A and Sviridov A P 2015 Applied Mechanics and Materials 698 374-377
[28] Hadzhieva O G, Illarionov A G and Popov A A 2012 Titan 4 21-26
[29] Meyers M A, Mishra A and Benson D J 2006 Progr. Mater. Sci. 51 427-556
[30] Zherebtsov S V, Salishchev G A, Galeyev R M, Valiakhmetov O R, Mironov S Yu and Semiatin S L 2004 Scripta Mater. 51 1147-1151
[31] Lutjering G and Williams J C 2007 In: Titanium (Springer-Verlag, Berlin/Heidelberg)
[32] Filimonov V Yu, Sitnikov A A, Afanas’ev A V, Loginova M V, Yakovlev V I, Negodyaev A Z, Schreifer D V and Solov’ev V A 2014 International Journal of Self-Propagating High-Temperature Synthesis 23, 1 18-25
[33] Filimonov V Yu, Sytnikov A A, Yakovlev V I, Loginova M V, Afanasyev A V and Negodyaev A Z 2014 Applied Mechanics and Materials 621 71-76