Combustion Synthesis of Nanocomposite Powders Using Mechanocomposite-Precursors

Tatiana L. Talako[1], Andrei I. Letsko[1], Piotr A. Vitiaz[1], Tatiana Ph. Grigorieva[2], Nikolai Z. Lyakhov[2],

[1]Laboratory of New Materials & Technologies, Powder Metallurgy Institute, 220005, Minsk, Belarus

[2]Laboratory of Chemical Material Science, Institute of Solid State Chemistry and Mechanochemistry, 630128, Novosibirsk, Russia

1 Introduction

The processing of nanostructured materials attracts more attention in the recent years. It is connected with the fact, that nanometer size scale brings substantial changes into physicochemical and mechanical properties, in comparison with bulk properties. Mechanically activated self-propagating high-temperature synthesis (MASHS) has been intensively investigated in the last decades as a method for production of nanomaterials [1-3]. The process consists of a short duration high-energy ball milling step followed by a self-sustaining synthesis reaction. Previously, it was shown that usage of preliminary mechanical activation of charge mixtures can lead to significant changes in combustion parameters [1-3]. As a rule, chemical interaction rate in mechanically activated mixtures is significantly higher as compared with the non-activated. This fact together with the lowered combustion temperature leads to formation of nanostructured materials even in the high-exothermal systems. This paper describes characteristic features of combustion processes in metal-oxide powder mixtures with aluminothermal reactions when using nanocomposite-precursors formed as a result of mechanochemical interaction of the mixture components at the stage of mechanical activation (MA).

2 Experimental

The experiments were performed in the next systems, wt.%: 60.9 Fe + 26.6 Al + 12.5 Fe_2O_3 (the 1-st system) and 65.9 Fe + 26 Al + 8.1 Cr_2O_3 (the 2-nd system). The powders production procedure included two stages. On the first step (mechanochemical activation) nano-composite metal/oxide powders were produced. On the second step self-propagating high-temperature synthesis (SHS) proceeded in the precursors formed.

For preparing mechanocomposites AGO-2 planetary ball mills with water cooling were used (drum volume 250 cm^3; ball diameter 5 mm; ball charge 200g; sample weight 10 g; rotation speed of grinding drums around the common axis 1000 rpm) [4]. As-produced mechanocomposites were ignited by an electrically heated tungsten coil in the SHS reactor. Both mechanical treatment and the SHS were carried out in argon atmosphere. Combustion wave temperature and velocity were measured

Correspondence to: talako@tut.by

with chromel-alumel thermocouple of 0.2 mm diameter using external two-channel twenty four-digit ADC ADSC24-2T.

Structure evolution was investigated using D8 Advance High Resolution Diffractometer with DIFRAC^{plus} software, High Resolution Scanning Electron Microscope "Mira" and Transmission Electron Microscope JEM1000x.

3 Results and Discussion

The general chemical equations of the reactions can be written as follows: $Fe + Al + Fe_2O_3 \rightarrow FeAl + Al_2O_3$ (the 1-st system) and $Fe + Al + Cr_2O_3 \rightarrow FeAl(Cr) + Al_2O_3$ (the 2-nd system).

Structure evolution of materials during MA and the following SHS is presented on Fig. 1. XRD analysis reveals different levels of interaction between components of reaction mixture for two systems. In the first system no Fe_2O_3 diffraction peaks were found in the powder diffraction pattern after MA (Fig. 1 Left). There is just a small peak between 35 and 36° 2θ, which can not be interpreted undoubtly (within the limits of an error it can belong to both Al_2O_3 and Fe_3O_4). At the same time, Al_2O_3 diffraction peaks can be well identified in the electronogramme of the precursor [5]. These data allow us to assume that during mechanical activation step Fe_2O_3 is reduced with aluminum, forming Al_2O_3.

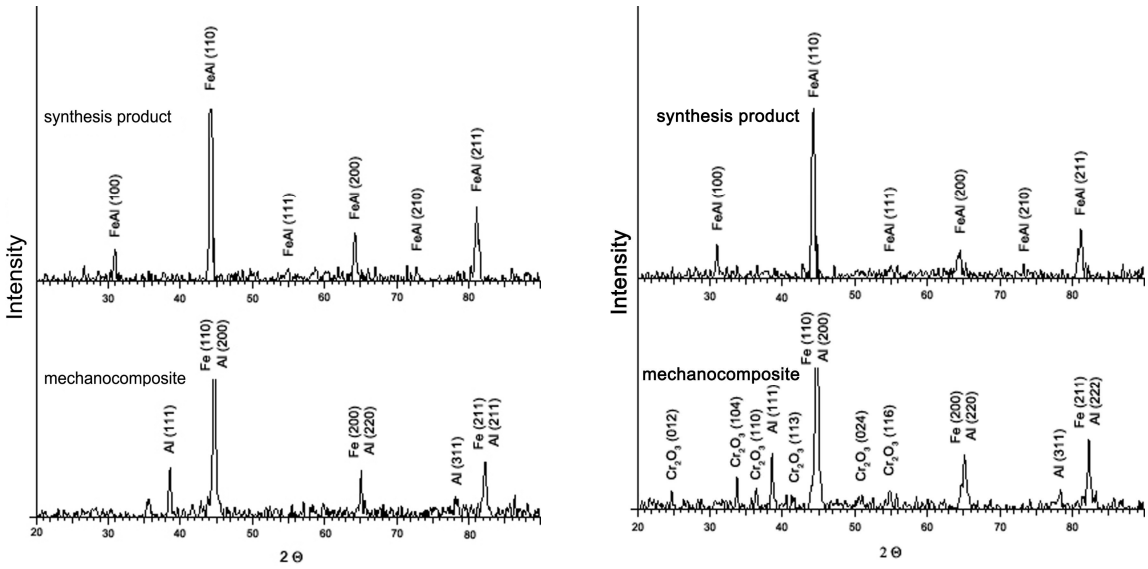

Figure 1. X-ray diffraction patterns of the mechanocomposite precursors and the SHS powders. Left: Fe-Al-Fe_2O_3 system. Right: Fe-Al-Cr_2O_3 system.

In the second system Cr_2O_3 diffraction peaks are still clearly defined after MA (see Fig. 1 Right). It is likely connected with the less oxide content in the charge mixture and with the lower exothermal effect of reaction $Cr_2O_3 + 2\ Al = Al_2O_3 + 2\ Cr$ as compared to aluminothermal reduction of Fe_2O_3 (~540 kJ/mol и ~840 kJ/mol, respectively). Though, TEM results testify presence of some content of nanosized alumina. Besides, according to results of Mossbauer spectroscopy investigations, about 2 % of Fe_2Al_5 is formed in material during MA [6].

In spite of the different levels of interaction between components in the mechanocomposites, combustion products in both SHS systems are quite similar (see Fig. 1). Main structural constituent of the both SHS powders is iron monoaluminide and corundum. According to DTA results, when heating at 10°/min, the precursors formed during MA in both systems interact via one-step reaction started at significantly lower temperature as compared with raw mixtures. SHS in both mechanocomposites

proceeds via the autooscillation mode. However, reaction dynamics is essentially different for the considered systems. Fig. 2 shows the combustion process thermogrammes.

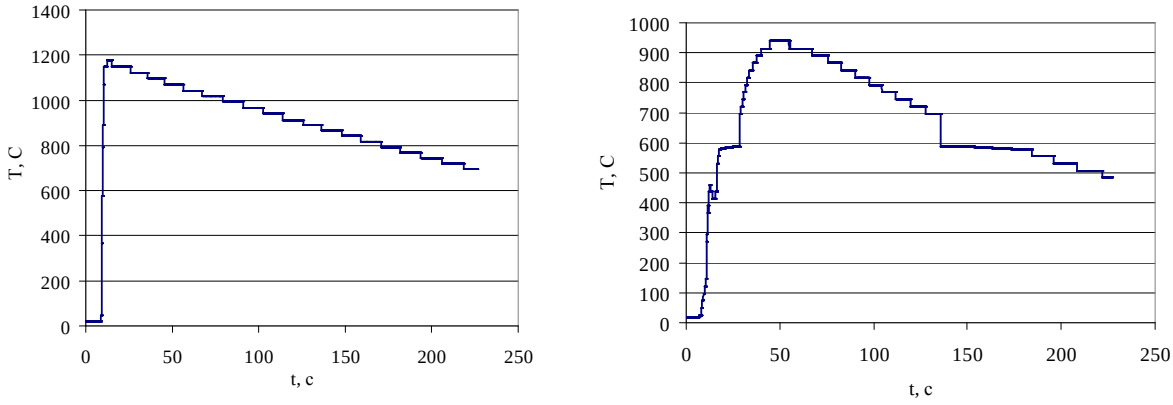

Figure 2. Thermogrammes of the combustion processes. Left: Fe-Al-Fe_2O_3 system. Right: Fe-Al-Cr_2O_3 system.

The initiation lag time for the 1-st system (Fe-Al-Fe_2O_3 raw mixture) is 9 s (Fig. 2 Left). Combustion wave temperature and velocity are 1150 °C и ≈0.3 mm/s, respectively. Chemical interaction completes in a few seconds followed by the relatively slow cooling down. Taking into account that combustion temperature is significantly higher than melting point of the least fusible eutectic in the system (T_{eu} Al-$FeAl_3$=652 °C), SHS with the intermediate molten layer evidently takes place.

Thermogramme of combustion in the 2-nd system (Fe-Al-Cr_2O_3) indicates more complicated behaviour with phase transformations in the zones of main heat evolution and after-burning (Fig. 2 Right). Combustion in the mechanocomposite is initiated in 7 s and runs with velocity ≈0.25 mm/s. As being expected, combustion temperature (942 °C) is some less than in the 1-st system. The isothermal stages are evidently connected with chromium dissolution in the aluminum melt (the first horizontal bend) and with the formation of the alloyed with chromium intermetallic $Fe_{0,70-x}Cr_xAl_{0,3}$ (x=0-0,2) [6] (the second horizontal bend).

It is interesting that both as-formed SHS powders completely inherit the precursor's structural morphology, being nanocomposites with partially amorphisated structure [5]. Absence of the deep sintering testifies the local appearance of the liquid phase at the contact areas and its low lifetime.

4 Conclusion

Usage of mechanocomposites as the SHS precursors allows formation of nanocomposite materials with complete inheritance of the precursor's structural morphology even in the systems with aluminothermal reactions. Complete reduction of oxides with aluminum at the stage of mechanical activation is not a necessary condition for the nanocomposite structure preservation. The last one is likely connected with the high rate of chemical reactions in the nanocomposite precursors and implementation of non-equilibrium phase formation mechanisms at the lowered temperatures.

Acknowledgements

This work was performed within of Suberian Branch of Russian Academy of Science and Belarussian Academy of Science integration programme N 98 "Mechanocomposites – precursors for the creation of materials with new properties".

References

[1] Bernard F, Gaffet E. (2001). Mechanical alloying in SHS research. Int. J. Self-Propag. High-Temp. Synth. 2: 109.

[2] Grigorieva T, Korchagin M, Lyakhov N. (2002). Combination of SHS and mechanochemical synthesis for nanopowder technologies. KONA. 20: 144.

[3] Levashov EA et al. (2007) Mechanoactivation of SHS system and Processes. Int. J. Self-Propag. High-Temp. Synth. 1: 46.

[4] Lyakhov NZ et al. (2005). Mechanochemically Synthesized Precursors for the Preparation of Intermetallic Compound/Oxide Nanocomposites by SHS. Doklady Chemistry. 405. Part 2: 255.

[5] Talaka T et al. (2007). Structure peculiarities of nanocomposite powder $Fe40Al/Al_2O_3$ produced by MASHS. Mater. Sci. Forum. 534-536: 1421.

[6] Kiseleva TYu et al. (2008). Redistribution of chromium atoms between components of nanocomposite intermetallis/oxide during synthesis. Moscow University Physics Bulletin. 4: 62.