Explosion Incited Magnesium Diboride Synthesis

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Abstract

For the first time MgB_2 and $MgCu_2$ were synthesized utilizing a dynamic loading technique. Particularly magnesium diboride is of great importance due to its superconducting properties. X-ray and electron microscopy analyses were carried out in order to characterize the products of the reaction. The method described can be used to produce pure MgB_2.

INTRODUCTION

There are several Mg–B derivatives known: MgB_2, MgB_4, MgB_6 and MgB_{12} [1, 2]. However, some of them lack stability. An interest towards these systems was stirred up in 2001, when it was found that MgB_2 had superconductivity at 40 K [3]. Nowadays MgB_2 is obtained in accordance with the following equation: $Mg + 2B = MgB_2$; the initial ratio of components and mode of thermal treatment [4] are varied during the process. The resulting product usually contains impurities of magnesium due to the time length of the process. The method of direct synthesis of MgB_2, utilized in a present work, made it possible to decrease the high temperature treatment period.

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EXPERIMENTAL

A thoroughly mixed compound in a stoichiometric proportion, consisting of two powders: magnesium powder (Mg96, metallic cuttings, 80–250 μm) and boron powder (B94 brand A, ≤1 μm), was loaded into a cylindrical ampoule. Parameters of the ampoule are as follows: outer diameter 14 mm, inner diameter 8 mm, length 100 mm. Gaseous atmosphere inside the reactor was the air. The ampoule was inserted inside the tubular explosive (ammonite–hexogen, mass ratio 1 : 1) charge. The diameter of the charge was kept constant in all experiments equal to 60 mm. Initiation of the explosion was made by means of a detonator situated at the upper central part of the ampoule.

Products of the explosion (with the speed equal to 3.6 and 5.5 km/s) made ampoule walls to contract, thus dynamic loading was developed. Pressure on the walls of the

ampoule, which is described as a pressure in a detonation wave, was assessed to be 3 and 7 GPa. These values exceed the strength of the reactor wall material; therefore the ampoule was deformed. The inward diameter of the ampoule diminished from 8 mm to about 5 mm, the reactor got pressurized and specific gravity of the reaction mass increased. For additional information refer to [5].

RESULTS AND DISCUSSION

Visual inspection of the cross-sections of the samples exposed to dynamical loading with pressure equal to 3 GPa revealed two distinct areas, *i.e.* the central part of the rod and its periphery. The latter part had a dark grey block structure, very similar to the initial mixture of powders. The former part (about 2 mm in diameter) had a structure of a sintered compact with uniform metallic lustre.

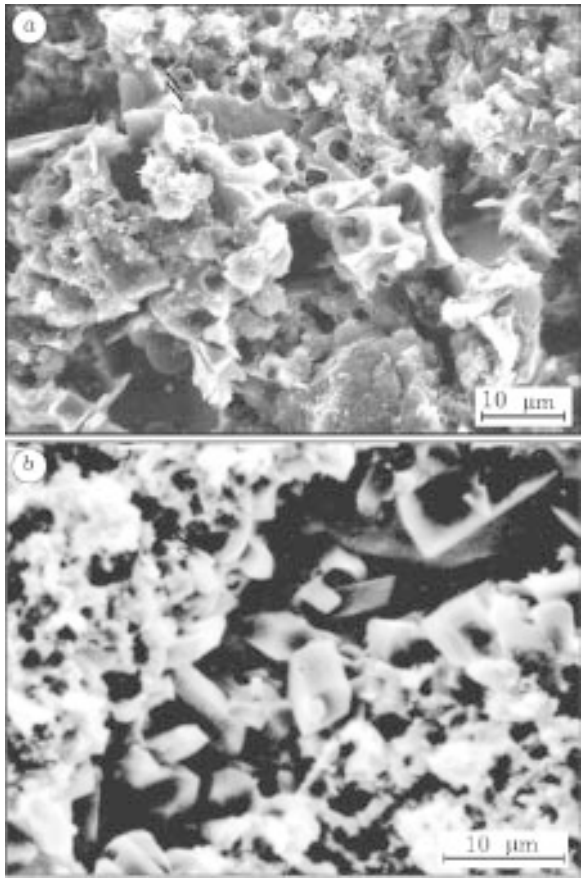

Fig. 1. Photomicrography of the central zone of the products after 3 GPa dynamic loading, yielded from: magnesium–boron system (*a*), magnesium–boron–copper system (*b*).

Investigations involving focused beam electronic microscope (JSM T-20) made it possible to discern at least three different areas in the cross-sections of the samples. These areas are periphery, central and intermediate part.

The peripheral zone had a block structure with low porosity and consisted of compacted initial mixture of reactants.

The central zone is of particular interest. It looks like a sintered mass with spherical voids in it (Fig. 1, *a*). These voids are thought to be formed during the process of the dynamic loading: particles of magnesium got melted and evaporated, leaving hollow space in the medium. Diffusion of gaseous Mg involved an exothermal reaction with boron.

The intermediate zone consisted of a blend of two macro-structures described above: unchanged reactants are mixed with the products of the process. X-ray analysis was carried out with the diffractometer DRON-3 (CuK_α radiation), with a graphite monochromator in order to lessen the noise level and to reveal fine structure of the spectra. The spectrum of the initial mixture showed magnesium signal only, as boron is an amorphous material. An identical type of the spectrum was obtained from the analysis of the peripheral zone after dynamic loading.

Though the central part was almost amorphous, three weak signals were detected. These signals could be referred to the MgB_2 phases (010), (011) and (110), hexagonal AlB_2 type, $a = 3.083$, $c = 3.521$. The percentage of these phases was estimated to be <10 %. No other borides were detected. The minor amount of magnesium was found to be among the products.

The samples produced from magnesium-boron powder after dynamic loading with the pressure of 7 GPa were studied visually and by means of metallography. The product of the reaction consisted of a hard, porous mass with metallic lustre, which can be regarded as an indication that the reaction took place.

Focused beam electronic microscopy applied to the specimen from the centre of the produced rod showed the presence of magnesium melts. This fact can be addressed in terms of the separation process. Massive magnesium particles got separated during the

progress of the blast waves towards the centre of the rod [6]. X-ray analysis also verified the presence of about 30 % magnesium in the central zone of the product mass. It can be inferred from these facts, that under such conditions separation process of magnesium preceded its exothermic reaction with boron.

Magnesium was not detected in the periphery of the reaction mass. The structure of the periphery (see Fig. 1, *a*) is similar to the structure of the bulk reaction product. According to the X-ray analysis, after 7 GPa dynamic loading, there was a twofold increase in the amount of MgB_2 in comparison to the 3 GPa dynamic loading results. The mixture, consisting of magnesium, boron powders with a 30 vol. % addition of copper powder (brand PM, 10–30 μm), was pounded and treated mechanochemically. Mechanochemical treatment was led for 1 min in an Ar atmosphere, with 60*g* acceleration in a planetary ball mill AGO-2 [7]. The resulting mixture was exposed to the explosion treatment (3 GPa) according to the technique described above.

The yielded product was similar to that obtained under the same conditions from the Cu-free mixture. A radial cross-section of the product revealed the presence of three discernible partitions: the periphery, the central (about 2 mm in diameter) and the intermediate zone, respectively.

X-ray analysis of the central zone specimen showed the presence of $MgCu_2$ and a small amount of copper. $MgCu_2$ crystals have characteristic rhombic structure as shown in Fig. 1, *b*. No magnesium diborides were detected. Thus, Cu can be regarded as an inhibitor towards explosion incited MgB_2 synthesis.

CONCLUSION

The results of the work are as follows:

– Dynamic loading synthesis of magnesium diboride was carried out for the first time.

– Analysis of the synthesis products showed there were no other magnesium borides or oxides present. Thus, the described technique of MgB_2 synthesis is a promising one. Pure magnesium diboride, obtained by means of the dynamic loading, can be used as a constituent of new composite materials with superconducting properties.

– For the first time $MgCu_2$ was synthesized from Mg–B–Cu mixture. It was found that $MgCu_2$ synthesis impeded magnesium borides production.

REFERENCES

1 G. V. Samsonov, V. P. Perminov, Magnidy, Nauk. Dumka, Kiev, 1971.
2 T. I. Serebryakova, V. A. Neronov, P. D. Peshev, Vysokotemperaturnye boridy, Metallurgiya, Moscow, 1991.
3 J. Nagamatsu, N. Nakagawa, T. Muranaka *et al.*, *Nature*, 410 (2001) 63.
4 A. L. Ivanovskiy, *Usp. Khim.*, 70, 9 (2001) 812.
5 V. I. Mali, T. S. Teslenko, *FGV*, 38, 4 (2002) 106.
6 N. A. Kostyukov, *PMTF*, 1 (1990) 84.
7 E. G. Avvakumov, Mekhanicheskiye metody aktivatsii khimicheskikh protsessov, Nauka, Novosibirsk, 1986.