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Explosive compaction and synthesis of MgB_2 superconductor using the powder in tube technique

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MgB_2 samples were prepared from Mg and B powders / flakes using the powder in tube method and consequent sintering in Argon atmosphere at 960 ^oC. Compaction of the Mg flakes and B powder resulted in a two phase alloy, with almost zero porosity and high hardness. XDR patterns illustrated that when Mg flakes and B powder were used as the precursor materials, formation of MgB_2 was not possible except for the border interface of Mg and B. Diffusion was not completed due to the low specific area. Contrary to that, the use of Mg and B fine powders lead to well packed superconductive MgB_2 samples.

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1. Introduction

Explosive consolidation of powders is a widely used cost-effective fabrication process due to its many advantages. Additionally, the process results in compacts with very good inter-particle bonding characteristics. On the other hand, isostatic compaction is a powder solidification process always followed by sintering in order to increase the material density and produce compacts of sufficient strength. The explosive compaction technique is based on the propagation of shock waves produced by a detonating explosive, transmitting the waves usually through a thin wall steel cylindrical container to the powder. This technique is known as the "powder in tube technique" [1]. This way, high shock pressures with a duration of a few microseconds can be produced, depending on the type and mass of the explosive as well as the way the shock waves are transmitted to the interior of the tube. Consolidation of the powder is caused by the container wall motion, accelerating towards the central axis of the tube, immediately after detonation. According to the stress wave propagation theory, an initial compressive stress wave is generated by the explosion, followed by a sequence of wave reflections as tensile and compressive pulses leading to collapse of the container and consequently of the tube content. The result of this process will be a dense compact provided that an adequate amount of energy is released by the explosion and transferred to the powder, while lack of energy will lead to a highly porous material.

Attempts to synthesise MgB_2 using self propagating high temperature synthesis (SHS method) have been reported indicating that near full density can be achieved [3], [4]. High pressure synthesis has also been used to prepare MgB_2 based bulk material with the addition of Ta in order to improve the superconductive properties of the material [5]. However the explosive compaction method to obtain a highly dense MgB_2 material from Mg and B powders has not yet been employed.

2. Experimental set-up

In this present work MgB_2 samples were prepared using the powder in tube method, whilst densification of the materials was carried out under explosive loading. The apparatus used for the explosive consolidation of the powders and flakes was designed in such a way so that the shock waves generated by the explosion are directly transmitted to the interior of the stainless steel container. As illustrated in Fig. 1, the explosive was placed in a cylindrical tube made of PVC and the whole arrangement was mounted on a specially designed 25mm thick steel plate, which acted as a shock wave absorber. The ends of the steel container were filled with a quantity of MgO powder and sealed with two plastic lids. MgO powder was used in order to avoid loosing part of the Mg or B material, since during the explosion process an amount of the tube content is blown into the air together with the plastic lids. The experiments were performed in an explosion proof room built on special foundations and the type of explosive used during the experiments was PETN with masses of 350.4 g, 370 g and 370.2 g.

The tube content was Mg flakes of 99.9% purity having a melting point of 651 ^oC and density of 1.75 g/cm^3, Mg powder of 98.5% purity, melting point of 651 ^oC and density of 1.74 g/cm^3 and B powder of 99% purity with a melting point of 2550 ^oC and density of 1.73 g/cm^3.

X-ray diffraction (XRD) patterns and scanning electron microscope (SEM) micrographs of the Mg – B and MgB_2 compacts were recorded before and after sintering. The XRD measurements were conducted with a Siemens D5000 diffractometer using nickel-filtered CuKα1 radiation (=1.5405 Å), 40 kV voltage and 30 mA current. In order to examine the material morphology, the explosive compaction products were also examined by scanning electron microscopy (SEM) using a Jeol 6100 Scanning Electron Microscope. Experimental conditions involved 20 kV accelerating voltage. Moreover ac susceptibility and hardness measurements were carried out using an ac susceptometer and a Leitz micro-hardness tester with a Vickers indenter, in order to determine the critical point of superconductivity and the material hardness respectively.

The sintering process was performed under argon atmosphere in a reactor chamber, constructed by resistance-heated quartz, with circular section. Finally the porosity of the Mg and B compacts was determined from photographs of various sections of the samples using a Leica model DMR microscope. The photographs were taken at 100X magnification.

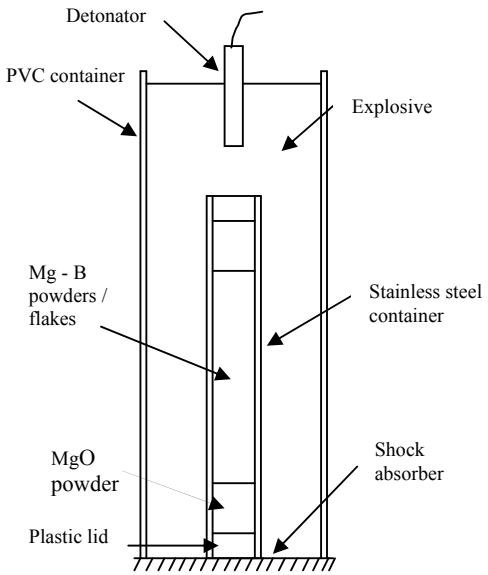

Fig. 1. Experimental set-up

3. Experimental results and discussion

The explosive compaction experiments involved two stainless steel containers filled with Mg flakes and B powder and a third one which included Mg and B powders after being blended in stoichiometric proportions 55.3% Mg and 44.7% B in a ball mill for 2 hours. A section of the sample after the explosive compaction is shown in Fig. 1, where the interface of steel and Mg-B compact is clearly visible. Scanning electron microscopy (SEM) and X-ray diffraction (XRD) illustrated that when Mg flakes and B powder were used as precursor materials the well packed cylinders did not form the MgB_2 superconducting phase. A cross section SEM micrograph, illustrating separate Mg and B phases of almost zero porosity, is given in the Fig. 3. The particle size of Mg flakes as determined from the SEM micrographs exceeds 100 μm, whilst B powder is a lot finer with an average particle size of 2 μm. XRD data are simply superposition of both Mg and B phases. This can be explained by the fact that the maximum temperature at the interior of the steel container during the explosive consolidation did not exceed 250 ^{o}C, which is significantly lower than the melting point of Mg.

Moreover, since it is a process of very small time duration, Mg and B could not react to form the MgB_2 phase. After the explosive compaction, the specimens were sectioned in several parts and sintered in Ar atmosphere. The compacts were heated up from room temperature up to 630 ^{o}C with a temperature rise of 5 K/min.

Fig. 2. Mg-B compact after consolidation in the stainless steel sheath.

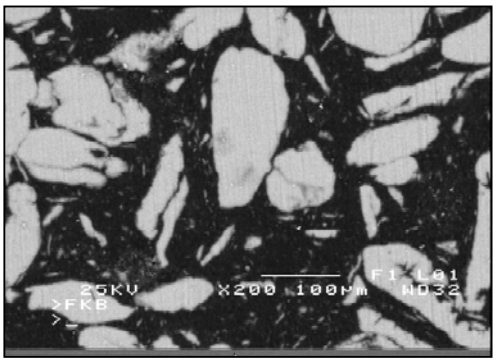

Fig. 3. SEM micrograph of Mg flakes and B powder compact before sintering.

The temperature remained constant at 630 ^{o}C for 30 minutes, and then slowly rose to 960 ^{o}C (with a rate of 1K/min) where it stayed for 90 minutes [2].

Then slow cooling was performed to 700 ^{o}C (1K/min) and at end the physical cool-down to ambient temperature.

Even after sintering, SEM images revealed the presence of two separate phases (Mg and B) since

diffusion was not completed due to the low specific area. MgB_2 is only present in very small quantities, at the border interface of Mg and B. Therefore it may be suggested that the reaction between B and Mg starts while the materials are still in solid state. This is illustrated in the XRD pattern of Fig. 5. The presence of MgO is attributed to partial oxidation of Mg flakes by air, which was trapped into the sheath during the explosive compaction.

The same technique was then followed using Mg and B fine powders and the samples were also undergone SEM and XRD structural characterization. The recorded results this time indicate the presence of superconducting phase, after heat treatment. Fig. 4 shows a cross section SEM micrograph illustrating MgB_2 compacted powder with almost zero porosity. No large size grains are detected since the micrograph corresponds to the sample prepared from Boron and Magnesium fine powders. The average grain size does not exceed 1 μm, illustrating the typical, expected, hexagonal structure of MgB_2. The MgB_2 superconducting phase is also cross-observed in the XRD response illustrated in Fig. 6. Indeed, ac susceptibility measurements as presented in Fig. 7 showing the real component of the material susceptibility X' as a function of temperature, reflected the presence of superconducting phase at 40.8 ^{o}K, which is a slightly higher transition temperature than what is usually reported for Fe sheathed MgB_2 superconductor, possibly due to impurities formed during the consolidation process.

The XRD results indicate that MgB_2 superconductor was directly synthesized by the mixed B and Mg powders but there are also additional components present, such as MgO and unreacted Mg in very small quantities.

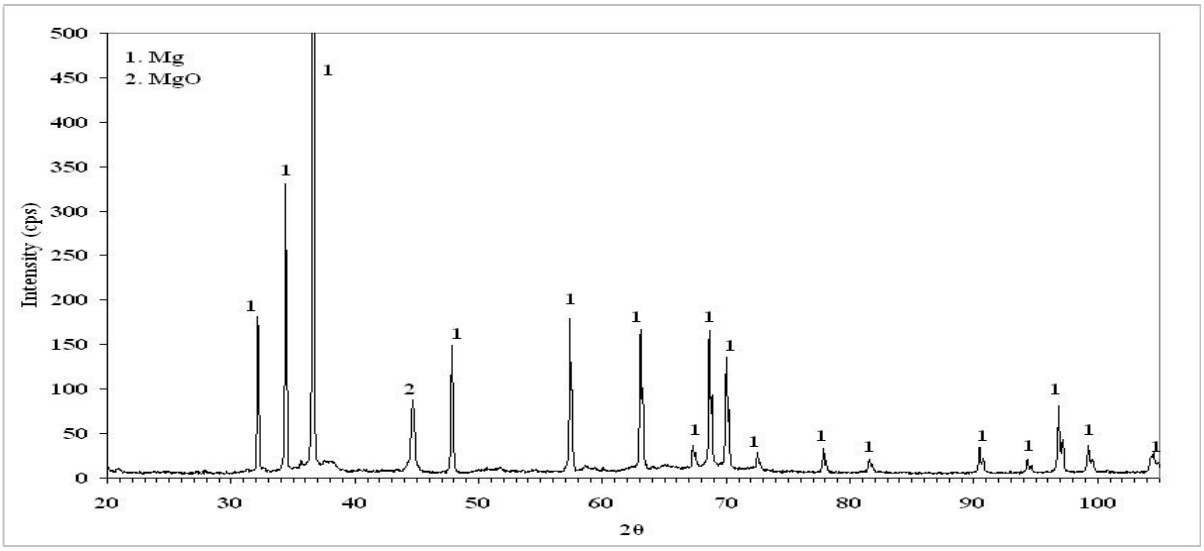

Fig. 5. XRD pattern of the sample produced from Mg flakes and B powders.

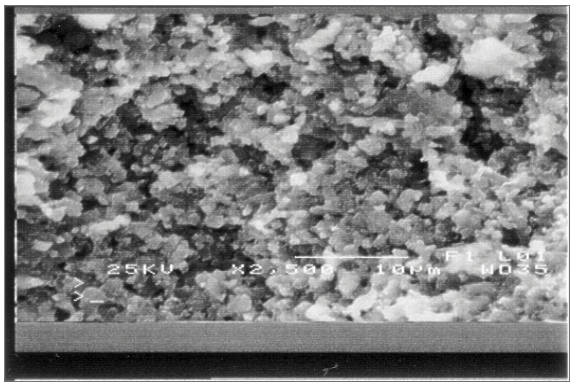

Fig. 4. SEM micrograph of MgB_2 powder compact after sintering

Synthesis of MgB_2 in this case was initialised during sintering, when melting of the surface layer of Mg and consequently diffusion of B into Mg started. Gradually, melting not only of the surface region but also of the interior of Mg grains was performed as temperature rose and diffusion was completed leading to a fine MgB_2 material as presented in the XRD patterns.

The microhardness range of the compact before sintering is between 39.30 HV and 57.14 HV from measurements carried out at locations mainly near the Mg – B interface which is very close to the actual hardness of Mg, while the corresponding microhardness values of MgB_2 are within the range of 1650 HV and 2700 HV. The recorded microhardness measurements are in good agreement with the properties of MgB_2 reported in the past [4].

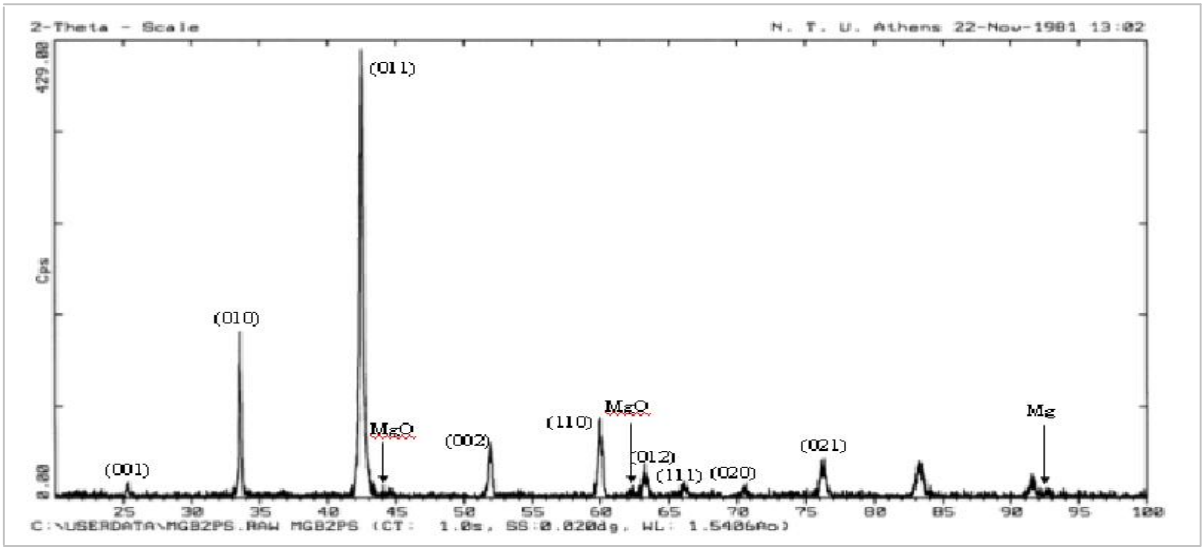

Fig. 6. XRD response of the compact produced from Mg and B powders, illustrating the presence of the MgB_2 phase

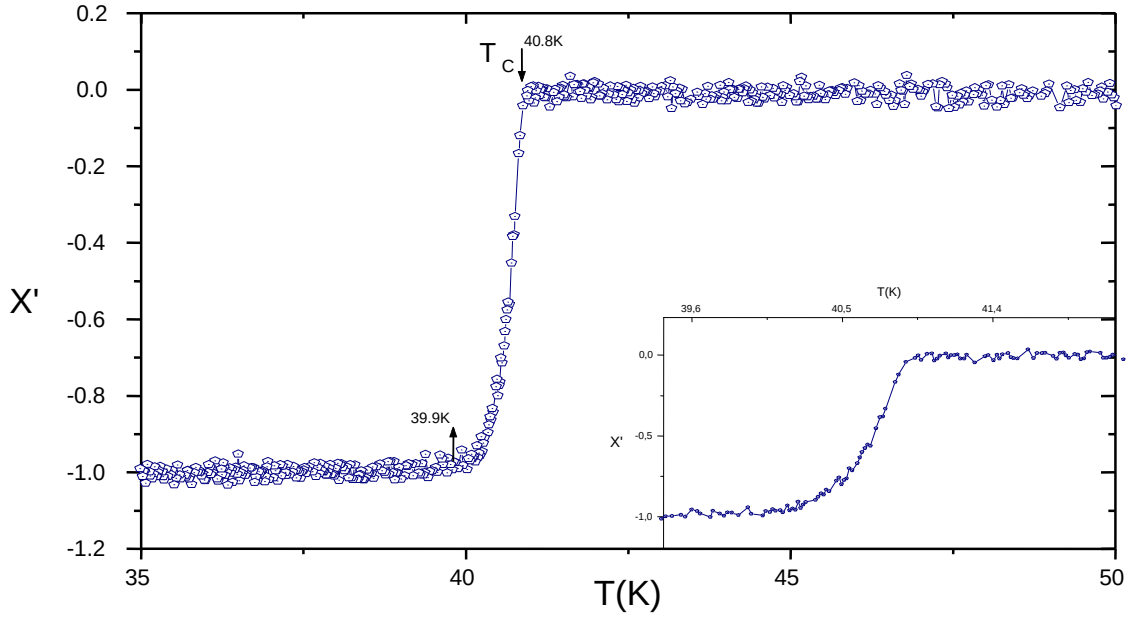

Fig. 7. AC susceptibility response showing a T_c of ~40K.

Porosity of the samples was also calculated and found to be nearly zero, indicating the importance of the explosive compaction technique when fabricating MgB_2 from Mg and B powders. Fig. 6 is a photograph of a section of the sample, at 100X magnification which was used in the porosity measurements.

Fig. 8. Microscope image of Mg and B compact at 100X magnification.

4. Conclusions

Stainless steel sheathed superconducting MgB_2 samples have been successfully fabricated from Mg and B fine powders using the powder in tube technique. Consolidation of the powders was performed under explosive loading and a consequent synthesis of MgB_2 was carried out by heating the compact in argon atmosphere up to 960 ^oC. Summarising the main features of the results reported, it may be concluded that the grain size of the precursor materials is crucial, since it affects diffusion of one material into the other. Synthesis of MgB_2 starts during sintering, whilst the powders are still in solid state and is located initially at the materials interface. The superconducting and mechanical properties of the material agree well with what is reported in text books.

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