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Compressive properties of pristine and SiC-Te-added MgB_2 powders, green compacts and spark-plasma-sintered bulks

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ABSTRACT

Pristine and (SiC + Te)-added MgB_2 powders, green and spark plasma sintered (SPS) compacts were investigated from the viewpoint of quasi-static and dynamic (Split-Hopkinson Pressure Bar, SHPB) compressive mechanical properties The amount of the additive (SiC + Te) was selected to be the optimum one for maximization of the superconducting functional parameters. Pristine and added MgB_2 show very similar compressive parameters (tan δ, fracture strength, Vickers hardness, others) and fragment size in the SHPB test. However, for the bulk SPSed samples the ratio of intergranular to transgranular fracturing changes, the first one being stronger in the added sample. This is reflected in the quasi-static K_{IC} that is higher for the added sample. Despite this result, sintered samples are brittle and have roughly similar fragmentation behavior as for brittle engineering ceramics. In the fragmentation process, the composite nature of our samples should be considered with a special focus on MgB_2 blocks (colonies) that show the major contribution to fracturing. The Glenn-Chudnovsky model of fracturing under dynamic load provides the closest values to our experimental fragment size data.

1. Introduction

MgB_2 is prized as a practical light-weight (relative density of 2.63 g/cm^3) superconductor [1] with a high potential of use in portable applications. MgB_2 was also proposed as a biomaterial [2] for fabrication of biodegradable devices and for other biomedical applications. As a superconductor or as a biomedical material, mechanical properties are important.

In general, mechanical properties of bulk MgB_2 are determined under static loads. They are shown to depend on the processing technology, additives and the quality of the sample. Namely, one has to consider the density, microstructure, phase composition, and the chemical composition of MgB_2 including substitutions, e.g. substitution of B by C. Indentation tests (Vickers hardness and fracture toughness), and room temperature bending strength tests are reported in refs. [3–14], respectively. Elastic constants of MgB_2 were calculated [15–17] or determined experimentally [18–20], and they are consistent with each other. The maximum measured values of Young modulus E, fracture toughness K_{IC}, Vickers hardness $HV_{>9.8N}$ and four-point bending strength $\sigma_{4\text{-point bending}}$ are 313 ± 9 GPa for pristine MgB_2 [10] (273 GPa from resonant ultrasound spectroscopy [18]) and ~240 GPa for C-substituted MgB_2 ($MgB_{2-x}C_x$, x = 0.1–0.3) [10], 4.4 ± 0.04 MPa m$^{0.5}$ for pristine and 7.6 ± 2 MPa m$^{0.5}$ for Ta-added MgB_2 [3], 14.94 ± 0.52 GPa [10], and 278 ± 43 MPa [12], respectively. The last two indicated values are for pristine MgB_2. It is worthy to note that even pristine MgB_2 bulk samples are usually composites containing secondary phases such as higher Mg-borides, Mg- and B-oxides.

MgB_2 is recognized as a material with a brittle behavior [10]. Previous paragraph indicates that quasi-static mechanical parameters, although inferior to typical light-weight and brittle engineering materials (such as e.g. Al_2O_3 with relative density of 3.95 g/cm^3), are still competitive and recommend MgB_2 as a potentially useful light-weight structural material. To target the already addressed applications or new ones, further exploration of the mechanical properties of MgB_2 is of high interest.

In this work we report compressive mechanical properties measured on pristine and SiC + Te co-added MgB_2 powders, green compacts and

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high density bulk samples prepared by spark plasma sintering (SPS). The added composition was selected considering our previous results and that critical current density is significantly higher than for the pristine sample [21]. SiC is a popular additive to MgB_2 [22] enhancing pinning. Addition of Te is also effectively providing pinning centers [23]. It also improves sintering processes towards a higher and more uniform density of the MgB_2 core from the powder-in-tube tapes [24]. Samples in powder form were subject to dynamic mechanical analysis (DMA). Quasi-static compressive tests were applied to green compacts. The two experiments are useful starting points in processing of sintered bulks and powder-in-tube tapes. We also present our results of quasi-static and dynamic (Split Hopkinson Pressure Bar, SHPB) tests on spark plasma sintered samples. Fractography and fragmentation details are discussed. It is worthy to note that for some superconducting applications, improvement of mechanical properties of MgB_2 tapes/wires and bulks is equally important for enhancement of superconducting functional parameters, but to the best of authors' knowledge, DMA on MgB_2 powders, quasi-static tests on green bodies and SHPB experiments on monolithic MgB_2 were not approached in literature.

2. Material and methods

Commercial powders (Alfa Aesar) were used: MgB_2 (99.5%, 1–2 μm), SiC nano-powder (Merck, 99% purity, 45 nm) and Te (C595090, Pierce Eurochemie B.V., 99.9999% purity, powder ground from a metal ingot). The MgB_2 powder contains, according to scanning electron microscopy (SEM), particles < 300 nm, agglomerated into larger aggregates up to 2–5 μm. The Te powder was composed of ~400 nm particles and large 2–10 μm blocks, as observed by SEM. The powders were mixed to obtain the composition $MgB_2(SiC)_{.025}Te_{.01}$. This composition was found to be the optimum one for maximization of the superconducting critical current density [21].

The pristine powder or the mixture with the indicated composition were loaded into a graphite die of ~2 cm inner diameter and sintered by SPS (FCT Systeme GmbH – HP D 5, Germany) for 3 min at 1150 °C. Vacuum in the SPS chamber was of 35 Pa. A uniaxial pressure of 95 MPa was applied on sample [7]. Our samples are denoted as follows: "MgB_2-pwd" for pristine MgB_2 powder, "MgB_2 + SiC + Te-pwd" for MgB_2 powder mixed with SiC and Te additions, "MgB_2-green" for pressed pristine powder, "MgB_2 + SiC + Te-green" for pressed MgB_2 + SiC + Te powder, "MgB_2-SPS" for pristine sintered bulk, and "MgB_2 + SiC + Te-SPS" for sintered MgB_2 with SiC and Te additions.

The apparent density (Table 1) of the sintered samples was measured by the Archimedes method. Relative densities were calculated as the ratio between the apparent and theoretical density [25], considering that samples contain MgB_2 (2.63 g/cm^3), MgB_4 (2.49 g/cm^3), MgO (3.58 g/cm^3), MgTe (3.86 g/cm^3), and Mg_2Si (1.988 g/cm^3).

The X-ray diffraction (XRD) spectra were taken with a Bruker-AXS D8 ADVANCE ($Cu_{k_{\alpha 1}}$-radiation, λ = 1.5406 Å) diffractometer. Rietveld refinement was performed and the weight fraction of each phase was determined with MAUD v.2.31 software [26]. The residual strain and the crystallite size (Table 1) by using the Williamson-Hall procedure [27] were calculated based on XRD data.

SEM images and energy dispersive X-ray spectroscopy (SEM-EDS) were observed with a Zeiss EVO50 microscope on surfaces of fractured samples.

A Quantum Design MPMS-7T was used to measure curves of magnetic moment versus temperature, $m(T)$, in zero field-cooling (ZFC) conditions. Pieces cut from the SPSed samples and fragments resulting after the SHPB dynamic impact test were investigated.

Dynamic mechanical analyses (DMA) of MgB_2 and MgB_2 + SiC + Te powders were performed on DMA Q800, TA Instruments, at room temperature (RT) with a force rate of 0.05 N/min, from 0 to 9 N, and from RT to 300 °C with a heating rate of 3 °C/min. Curves of strain and stiffness with time and temperature are presented in Fig. 1. Values of the compressive loss modulus E″ (viscous component) and storage modulus E′ (elastic component) were obtained and their ratio E″/E′ defines the loss tangent tan(δ) (where δ = phase lag between stress and strain).

Compression quasi-static experiments on green bodies (diameter = 10 mm, height = 10 mm) were performed on Instron 3382, with a compression rate of 0.5 mm/min. Green bulks were obtained for a load of 50 kN (637 MPa).

Bulk SPSed samples were cut into cylinders (diameter = 4.0 mm, height = 4.0 mm) by wire electrical discharge machining. Samples were subject to uniaxial compression (Instron 5569) with a rate of 0.24 mm/min. The compression process of the samples was captured by a JAI (JAI Ltd., Japan) BM-500 GE high resolution camera to accurately track and measure the strain history.

Vickers hardness (*HV*) was measured on a polished surface of the SPSed sample for a load of 2 kgf (19.6 N) using a CV-400DTS Micro Hardness Tester. We applied the standard procedure according to ASTM C 1327-03. Ten measurements were performed with a dwell time of 10 s each. The average hardness and values of the standard deviation were determined (Table 1). Based on Vickers hardness and the induced cracks, we calculated the static fracture toughness K_{IC} (Table 1) according to ref. [28]:

$$K_{IC} = 0.048 \bullet \left(\frac{l}{a}\right)^{-\frac{1}{2}} \bullet \left(\frac{HV}{E \bullet \Phi}\right)^{-\frac{2}{5}} \bullet \left(\frac{HV \bullet a^{\frac{1}{2}}}{\Phi}\right) \quad (1)$$

where: K_{IC} = fracture toughness (MPa·m$^{1/2}$), ℓ = crack length (μm), a = indent half-diagonal (μm), HV = Vickers hardness (GPa), E = Young's modulus (GPa), Φ = constraint factor (≈ 3). Berkovich hardness was measured with an Agilent G200 nano indenter (Table 1).

Uniaxial dynamic compression test on SPSed samples (diameter = 4.0 mm, height = 4.0 mm) was conducted using a Split-Hopkinson Pressure Bar (SHPB), designed at NTU, Singapore. The apparatus consists of 20 mm diameter YAG300 maraging steel striker (length 400 mm), incident and transmitted (both 1200 mm long) bars. The incident and transmitted bars were instrumented with TML strain gauges (Tokyo Sokki Kenkyujo Co. Ltd., Japan, gauge factor of 2.11). Signals from strain gauges were used to calculate the stress and strain history based on the one-dimensional elastic bar wave theory for a pulse propagating in a uniform bar (SHPB theory) as described in ref. [29]. Hardened high strength steel plates with impedance matching that of the bars were sandwiched between the bars and specimens to avoid the indentation of the bars [30,31]. In the analysis of the SHPB test, the longitudinal wave velocity of the maraging steel bars is $c_0 = \sqrt{E_0/\rho_0} = 4786 m \bullet s^{-1}$, where Young's modulus E_0 = 184 GPa and density ρ_0 = 8030 kg m^{-3}.

The dimensions of resulted fragments in the SHPB test were measured (width and length) using Gimp 2.8 and ImageJ 1.6 software on SEM images. Statistical analysis was applied on 926 and 815 fragments that resulted from the SHPB impact test of the samples MgB_2-SPS, and MgB_2 + SiC + Te-SPS, respectively. Statistical toolbox of MATLAB was used to calculate the lognormal distribution of the fragment sizes resulting after SHPB based on Eq. (2)

$$y = \frac{1}{\sigma \bullet x \bullet \sqrt{2 \bullet \pi}} e^{\frac{-(\ln x - \mu)^2}{2 \bullet \sigma^2}} \quad (2)$$

where y is the *probability density function* (PDF), x is the random variable (here width or length), σ and μ are the standard variation and the mean value of ln x.

3. Results and discussion

3.1. DMA on powders

Curves of DMA for samples MgB_2-pwd and MgB_2 + SiC + Te-pwd are presented in Fig. 1. Strain and stiffness curves vs. time and temperature show some differences between pristine and added sample.