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Effects of oxide particle addition on superconductivity in nanocrystalline MgB_2 bulk samples

Olaf Perner [a,b,*], Wolfgang Häßler [a,b], Jürgen Eckert [a,b,c], Claus Fischer [a,b], Christine Mickel [a,b], Günter Fuchs [a,b], Bernhard Holzapfel [a,b], Ludwig Schultz [a,b]

[a] *IFW Dresden, Institute for Metallic Materials, P.O. Box 270116, D-01171 Dresden, Germany*
[b] *Dresden University of Technology, Department of Physics, Institute for Physics of Solids, D-01062 Dresden, Germany*
[c] *Technische Universität Darmstadt, FB 11 Material- und Geowissenschaften, FG Physikalische Metallkunde, Petersenstraße 23, D-64287 Darmstadt, Germany*

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Abstract

In order to improve the extrinsic superconducting properties of MgB_2 a combination of nanocrystalline microstructure and particle doping was studied with regard to the enhancement of flux pinning. For comparison, MgO and SiO_2 particles were separately introduced into the MgB_2 matrix by the in situ preparation technique of mechanical alloying. The oxides do not affect the formation of MgB_2 but influence characteristically the critical current density J_c and the irreversibility field H_{irr} in hot pressed bulk samples due to a change of microstructure and lattice constants. Optimal doping levels are 5 wt.% for MgO and 2 wt.% for SiO_2 addition, which lead to improved J_c values of 1.3×10^6 and 1.2×10^6 A/cm^2 in self-field at 7.5 K, respectively, but do not affect the critical temperature T_c. Microstructural investigations revealed a nanocrystalline MgB_2 matrix with homogeneously distributed impurity particles.

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1. Introduction

One main focus in the investigation of superconductivity in MgB_2, which was discovered by Nagamatsu et al. [1], has been the improvement

* Corresponding author. Address: IFW Dresden, Institute for Metallic Materials, P.O. Box 270116, D-01171 Dresden, Germany. Tel.: +49 351 46 59 640; fax: +49 351 46 59 541.
E-mail address: o.perner@ifw-dresden.de (O. Perner).

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of the properties of this material for potential applications like magnets, cables and superconducting circuits [2,3]. The enhancement of the critical current density as well as the irreversibility field can be achieved by the introduction of pinning centers [4]. For this, different approaches like defect creation by high energy radiation and grain refinement as well as element doping and nanoparticle addition [4,5] were investigated. Dou et al. [6] have introduced the idea of SiC addition, which leads to enhancement of J_c and H_{irr} while leaving the critical temperature T_c almost unchanged.

In a model of two-band scattering in superconductors like MgB_2, Gurevich [7] describes the possibility of H_{c2} enhancement by increasing the normal state resistivity as well as by increasing the intraband scattering rates due to substitution on Mg and B sites without affecting T_c.

The effect of MgO particles present in the MgB_2 microstructure was already investigated by different groups [8–10]. Kovác et al. [8] have shown that an enhancement of J_c and H_{irr} can be achieved while leaving T_c almost unchanged. But the improvement of pinning worsens the inter-grain connectivity. Riu et al. [11] studied the influence of SiO_2 on the properties of MgB_2 and found an optimum concentration of 7 at.% for J_c enhancement. Si is doped into the MgB_2 lattice cell whereas the formation of Mg_2Si particles leads to nanocomposite material. But the grain size of these particles of about 50 nm is too large to enable an optimum pinning efficiency, which is expected for 10–20 nm sized pinning centers because the coherence length in MgB_2 is in the range of 6–7 nm [12].

A combination of different mechanisms to enhance the pinning properties of MgB_2 was the aim of the present work and will be described in detail in the following. The mechanical alloying technique was used for the in situ preparation of MgB_2 starting from magnesium and boron powders with the addition of different fractions of MgO and SiO_2 particles. The idea behind this was to fabricate a nanocrystalline MgB_2 microstructure as well as to achieve a grain size reduction of the oxide particles and their effective incorporation into the MgB_2 matrix. Whereas MgO is always found as a secondary phase in MgB_2, which forms due to the presence of small amounts of oxygen in the working atmosphere [13], SiO_2 can be regarded as a real impurity phase whose influence on the superconductivity in MgB_2 is more complex [11].

The phase formation of MgB_2 as well as the superconducting properties in dependence of the microstructure were studied. In fact, we find a different behaviour of MgO and SiO_2 addition in dependence on the doping content on the properties of MgB_2. Slight additions of these impurities were found to improve the pinning properties without changing T_c. In contrast, larger amounts of MgO and SiO_2 deteriorate the superconducting properties due to a change of the microstructure and the grain connectivity.

2. Experimental

The preparation of MgB_2 using the mechanical alloying (MA) technique was carried out with elementary amorphous boron (1 μm particle size, 99.99% purity, Alfa Aesar) and magnesium (250 μm maximum particle size, 99.9% purity, Goodfellow) powders in the stoichiometric composition. Furthermore, MgO (45 μm maximum particle size, 99.5% purity, Alfa Aesar) and SiO_2 (45 μm maximum particle size, 99.8% purity, Chempur) powders were added resulting in a phase fraction between 2–10 wt.% and 2–20 wt.%, respectively.

Mechanical alloying was done in a Retsch PM 400 planetary ball mill with milling tools made of tungsten carbide (WC) in a highly purified argon atmosphere with the following parameters: 50 h milling time, 250 rpm rotational speed, ball-to-powder mass ratio of 36:1. Further details are described elsewhere [13,14].

In order to receive highly densified bulk samples the as-milled and partially reacted MgB_2 powder was hot pressed in a vacuum chamber at a temperature of 973 K under a pressure of 500 MPa for 10 min (for further details, see [13,14]).

Phase analysis was performed using a Philips X'Pert PW 3040 X-ray diffractometer with Co K_α radiation. Rietveld refinement [15] of the obtained X-ray diffraction patterns using MgB_2, Mg, MgO, SiO_2, Mg_2Si and WC peaks allowed to determine