Flux Pinning Optimization of MgB_2 Bulk Samples Prepared Using High-Energy Ball Milling and Addition of TaB_2

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***Abstract*—MgB_2 is considered to be an important conductor for applications. Optimizing flux pinning in these conductors can improve their critical currents. Doping can influence flux pinning efficiency and grain connectivity, and also affect the resistivity, upper critical field and critical temperature. This study was designed to attempt the doping of MgB_2 on the Mg sites with metal-diborides using high-energy ball milling. MgB_2 samples were prepared by milling pre-reacted MgB_2 and TaB_2 powders using a Spex 8000M mill with WC jars and balls in a nitrogen-filled glove box. The mixing concentration in $(Mg_{1-x}Ta_x)B_2$ was up to $x = 0.10$. Samples were removed from the WC jars after milling times up to 4000 minutes and formed into pellets using cold isostatic pressing. The pellets were heat treated in a hot isostatic press (HIP) at 1000°C under a pressure of 30 kpsi for 24 hours. The influence that milling time and TaB_2 addition had on the microstructure and the resulting superconducting properties of TaB_2-added MgB_2 is discussed. Improvement of J_c at high magnetic fields and of pinning could be obtained in milled samples with added TaB_2. The sample with added $5at.\%TaB_2$ and milled for 300 minutes showed values of $J_c \sim 7 \times 10^5$ A/cm^2 and $F_p \sim 14$ GN/m^3 at 2 T, 4.2 K. The milled and TaB_2-mixed samples showed higher values of $\mu_0 H_{irr}$ than the unmilled-unmixed sample.**

***Index Terms*—Addition of diborides, high-energy ball milling, MgB_2, microstructural and superconducting characterization.**

I. Introduction

MAGNESIUM diboride (MgB_2) has already been demonstrated for low-field MRI magnets and has the potential for higher field applications [1]. The high critical temperature (T_c) and potentially high upper critical field (H_{c2}) of MgB_2 creates the possibility that this material can replace Nb-based superconductors in several of these applications [2]–[4]. MgB_2 wires and bulks can be produced from powders in Cu-alloy tubes, using the in-situ or the ex-situ method, many of them adapted from the Nb_3Sn production technology [5], [6]. The critical current densities are approaching those of NbTi at 8 T and 4.2 K, and it has the potential to be further improved.

MgB_2 has low raw material costs and can operate at higher temperatures than Nb superconductors using cryogenic refrigerators [5], [6]. Unlike the HTS superconductors, MgB_2 does not have problems of grain boundary weak links [7].

However, further optimization of magnetic flux pinning in these conductors is of vital importance for the improvement of their critical currents [8]. As a granular superconductor, it is also important to improve densification and grain boundary connectivity. The processing scheme and the introduction of defects and doping in the superconducting matrix can influence the flux pinning efficiency and grain connectivity, also affecting the stoichiometry of the superconducting phase, changing its resistivity, H_{c2} and T_c, important properties and characteristics for practical applications [9], [10].

The present work shows the use of high-energy ball milling to prepare MgB_2 bulk superconductors. This study was designed to attempt the doping of MgB_2 on the Mg sites with metal-diborides using high-energy ball milling. The MgB_2 samples were prepared by milling pre-reacted MgB_2 and TaB_2 powders (ex-situ method). The samples were fully prepared in a glove box with a nitrogen atmosphere to decrease the possibility of contamination from O_2 and H_2O, and they were compacted using cold isostatic pressing (CIP). The pellets were heat treated using hot isostatic pressing (HIP) to densify and sinter the $MgB_2 + TaB_2$ powder. Microstructural and superconducting characterization of samples were performed to determine the influence that milling, TaB_2-addition and heat treatment had on their properties and characteristics.

II. Experimental Procedures

MgB_2 bulk samples were prepared using pre-reacted MgB_2 and TaB_2 Alfa-Aesar powders, −325 mesh. The powders were milled using a SPEX 8000M mill with WC jar and balls in a glove box with a nitrogen atmosphere. The mixing concentration in $(Mg_{1-x}Ta_x)B_2$ was up to $x = 0.10$.

Samples were removed from the WC jars after milling times ranging up to 4000 minutes and formed into pellets using cold isostatic pressing (CIP) under a pressure of 30,000 psi. The samples were kept under nitrogen atmosphere during all the milling and cold-compaction processes.

After CIP, the pellets were sealed under vacuum in stainless steel tubes and heat treated in a hot isostatic press (HIP) at 1000°C under a pressure of 30 kpsi for 24 hours. One sample

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was CIPped and HIPped without the milling procedure and without TaB_2 addition (MgB_2 raw unmilled).

Slices were cut from the HIPped samples milled for 300 minutes and for 4000 minutes. Microstructural characterization was performed using X-ray diffractometry (XRD) with $Cu - K\alpha$ radiation in a STOE diffractometer, just after milling and after the HIP heat treatment. The crystal structure refinement of all samples was performed using the PowderCell Rietveld refinement software (freeware) [11]. It was possible to extract an estimate of the lattice parameters, average particle size and strain.

The superconducting characterization of the samples was performed using a 9 T Quantum Design Physical Properties Measurement System (PPMS). The critical temperature T_c was defined as the onset of the ZFC transition from the DC magnetization versus temperature curves. DC magnetization loops versus applied magnetic field at 4.2 K were used to extract the $J_c(\mathrm{H}, \mathrm{T} = 4.2\ \mathrm{K})$ vs. $\mu_0 \mathrm{H}$ curves using

$$J_c = M \cdot 12b/(3bd - d^2) \tag{1}$$

where b and d are the width and thickness of the rectangular-shaped section extracted from the samples, following the Bean model [12]. The volumetric pinning force $F_p(\mathrm{H}, \mathrm{T} = 4.2\ \mathrm{K})$ vs. $\mu_0 H$ curves were calculated from this magnetization J_c.

III. Results and Discussion

A. Microstructural Characterization

The microstructural characterization of MgB_2 without TaB_2 addition (unmixed) was presented elsewhere [13]. It was found that the as-received MgB_2 contained around 1.6 wt.% MgO and that the MgO content increased to 7–8 wt.% after the HIP heat treatment for all milled samples. The MgB_2 average particle size decreased with milling time and contamination by WC was found in the samples after long milling times.

Table I shows the structural refinement results obtained for samples milled for 300 and 4000 minutes, before and after HIP. In general, the XRD patterns of the milled samples before HIPping were broad and of low intensity. We assume the samples still contained MgO even though the MgO peaks could not be seen in the milled unHIPped samples. As seen for the unmixed samples, the data in Table I show an expected decrease in particle size and increase in strain compared to the as-received MgB_2. The MgB_2 lattice parameters in the milled TaB_2-added samples remained almost the same as those for the unmilled unmixed materials. After HIP, the samples always contained some additional MgO and the MgB_2 a lattice parameters were almost unchanged. The MgB_2 average particle size always increased and the strain always decreased for the HIPped samples compared to the milled samples. The strain behavior could be expected due to the grain growth and energy release during HIPping.

The samples milled for 4000 minutes did not show MgO before HIP, but showed the presence of WC. This is an important result that shows the contamination from the milling media in the powder after long milling times. This is a general result that is applicable to any MgB_2 prepared using WC milling media [13], [14].

TABLE I
Structural Refinement for the MgB_2 Samples

Sample	Composition	a (Å)	c (Å)	Average particle size (nm)	Strain
MgB_2 raw unmilled	98.4wt.%MgB_2	3.0880	3.5241	97.0	0.001855
	1.6wt.%MgO	4.2342	-	-	-
MgB_2 raw unmilled	92.2wt.%MgB_2	3.0844	3.5262	73.0	0.001098
(after HIP)	7.8wt.%MgO	4.2242	-	19.5	0.001600
MgB_2 milled 300 min	100wt.%MgB_2	3.0888	3.5262	16.3	0.006117
MgB_2 milled 300 min	92.7wt.%MgB_2	3.0875	3.5299	99.8	0.003385
(after HIP)	7.3wt.%MgO	4.2255	-	56.0	0.002563
MgB_2 milled 4000	96.6wt.%MgB_2	3.0788	3.5355	8.1	0.006900
min	3.4wt.%WC	2.9041	2.8403	12.0	0.003908
MgB_2 milled 4000	88.7wt.%MgB_2	3.0863	3.5299	113.1	0.003357
min (after HIP)	11.3wt.%MgO	4.2259	-	45.4	0.003843
MgB_2+2at.%TaB_2	98.5wt.%MgB_2	3.0879	3.5259	12.8	0.003571
milled 300 min	1.5wt.%TaB_2	3.1007	3.2294	45.2	0.001691
MgB_2+2at.%TaB_2	86.5wt.%MgB_2	3.0802	3.5214	57.9	0.000812
milled 300 min	6.0wt.%TaB_2	3.0906	3.2339	44.8	0.000538
(after HIP)	7.5wt.%MgO	4.2116	-	34.1	0.000703
MgB_2+2at.%TaB_2	90.6wt.%MgB_2	3.0785	3.5194	7.0	0.006324
milled 4000 min	7.5wt.%TaB_2	3.0995	3.2311	21.2	0.001279
	1.8wt.%WC	2.9072	2.8416	27.0	0.004655
MgB_2+2at.%TaB_2	83.5wt.%MgB_2	3.0827	3.5244	32.3	0.000950
milled 4000 min	5.8wt.%TaB_2	3.0950	3.2416	22.4	0.001415
(after HIP)	10.7wt.%MgO	4.2179	-	31.0	0.000935
MgB_2+5at.%TaB_2	84.6wt.%MgB_2	3.0891	3.5342	12.2	0.003584
milled 300 min	15.4wt.%TaB_2	3.1040	3.2304	44.2	0.001686
MgB_2+5at.%TaB_2	77.3wt.%MgB_2	3.0871	3.5282	14.4	0.002285
milled 300 min	15.1wt.%TaB_2	3.0939	3.2457	43.9	0.000286
(after HIP)	7.6wt.%MgO	4.2146	-	38.3	0.001764
MgB_2+5at.%TaB_2	80.2wt.%MgB_2	3.0770	3.5178	8.0	0.006865
milled 4000 min	14.6wt.%TaB_2	3.1012	3.2346	14.4	0.001298
	5.1wt.%WC	2.9071	2.8468	21.7	0.007225
MgB_2+5at.%TaB_2	70.9wt.%MgB_2	3.0786	3.5273	27.5	0.000208
milled 4000 min	13.3wt.%TaB_2	3.0945	3.2468	16.4	0.001850
(after HIP)	15.8wt.%MgO	4.2193	-	27.3	0.003804

The structural refinements of the HIPped samples milled for 4000 minutes show a decrease in the particle size and an increase in the strain compared to the raw material, the unmixed samples, and the samples milled for 300 minutes before HIPping. The a lattice parameter of the 4000 minute milled unHIPped samples (with and without TaB_2) increased with HIPping. In contrast, the a lattice parameter of the unmilled and 300 minute samples (with and without TaB_2) were essentially identical, equivalent to the 4000 minute HIPped sample. This is possibly a direct result of high-energy ball milling for long periods of time. After HIP, the XRD patterns did not show WC but did show MgO. The WC found in the milled powder before HIPping is due to the low crystallinity and small particle size of MgB_2, which gave broad and weak X-ray peaks, whereas the dense, crystalline WC, gave sharp and strong peaks. After HIPping, the MgB_2 grains became more crystalline, resulting in strong and sharp XRD peaks that dominated the XRD pattern. However, some WC contamination is still present in the samples.

The TaB_2 average particle size did not change after HIPping, when compared to the milled samples. This is an indication that the milling created a composite consisting of a MgB_2 matrix with finely dispersed TaB_2 particles. This is an important result to analyze the transport and pinning characteristics of these samples.

In general, the results show the particle size decreased and the strain increased with increased milling time for the mixed samples. HIPping increased particle size and decreased strain.

TABLE II
CRITICAL TEMPERATURE T_c FOR THE MgB_2 SAMPLES. ALL SAMPLES AFTER HIPPING

Sample	T_c (K) Onset magnetization
MgB_2 raw unmilled-unmixed	38.5
MgB_2 milled 300 min	37.5
MgB_2 milled 4000 min	33.9
MgB_2+2at.%TaB_2 milled 300 min	37.5
MgB_2+2at.%TaB_2 milled 4000 min	34.5
MgB_2+5at.%TaB_2 milled 300 min	37.6
MgB_2+5at.%TaB_2 milled 4000 min	29.5
MgB_2+10at.%TaB_2 milled 300 min	37.5
MgB_2+10at.%TaB_2 milled 4000 min	33.0

B. Superconducting Characterization

The onset temperature of the superconducting transition, defined as T_c, was reduced by the high-energy ball milling. Table II shows the T_c values extracted from DC magnetization measurements. The milling always reduced T_c, even after short milling times, when compared to the unmilled sample. It can be seen that the samples milled for 300 minutes showed almost the same values of T_c, for all TaB_2-adding concentration. However, the samples showed different values of T_c after long milling times, depending on mixing content.

Fig. 1 shows the magnetization J_c as a function of applied magnetic field at 4.2 K for the HIPped samples. Fig. 1(a) shows the comparison for the samples mixed with $2\text{at.}\%TaB_2$. At very low magnetic fields, the unmilled-unmixed sample had the highest values of J_c. It was found that milling increases J_c at high magnetic fields compared to the unmilled MgB_2. The benefits resulting from milling are at higher fields, most probably due to milling-induced effects resulting in increased electron scattering and $\mu_0 H_{c2}$. This explains the increase of J_c for these samples at high fields and the increase in the irreversibility field $\mu_0 H_{irr}$ of the milled samples, which is extracted from the curves in Fig. 1 by extrapolating to $J_c = 100\ \text{A/cm}^2$. It can be seen that the sample unmixed and milled for 300 minutes showed the highest values of J_c for almost the entire field range. The unmixed sample milled for 4000 minutes seems to have higher values of $\mu_0 H_{irr}$.

Fig. 1(b) shows the comparison for the samples mixed with $5\text{at.}\%TaB_2$. In general, mixing and milling increased J_c for all samples in magnetic fields above 3 T. Milling effectively increased $\mu_0 H_{irr}$. Mixing with $5\text{at.}\%TaB_2$ and milling for short times (300 minutes) increased J_c at low magnetic fields compared to the unmilled and to the unmixed samples. For all samples, milling was important to improve J_c and $\mu_0 H_{irr}$. This could optimize the application of these materials.

Fig. 1(c) shows the comparison for the samples mixed with $10\text{at.}\%TaB_2$. Again, as an average behavior, mixing and milling increased J_c for all samples in magnetic fields above 3 T. Milling was effective to increase $\mu_0 H_{irr}$. Mixing with $10\text{at.}\%TaB_2$ and milling for long times (4000 minutes) increased J_c at magnetic fields above 8 T, compared to the unmilled and to the unmixed samples. For all samples, milling was important to improve J_c and $\mu_0 H_{irr}$. However, mixing with $10\text{at.}\%TaB_2$ and milling for short times did not improve the behavior compared to the unmilled-unmixed sample.

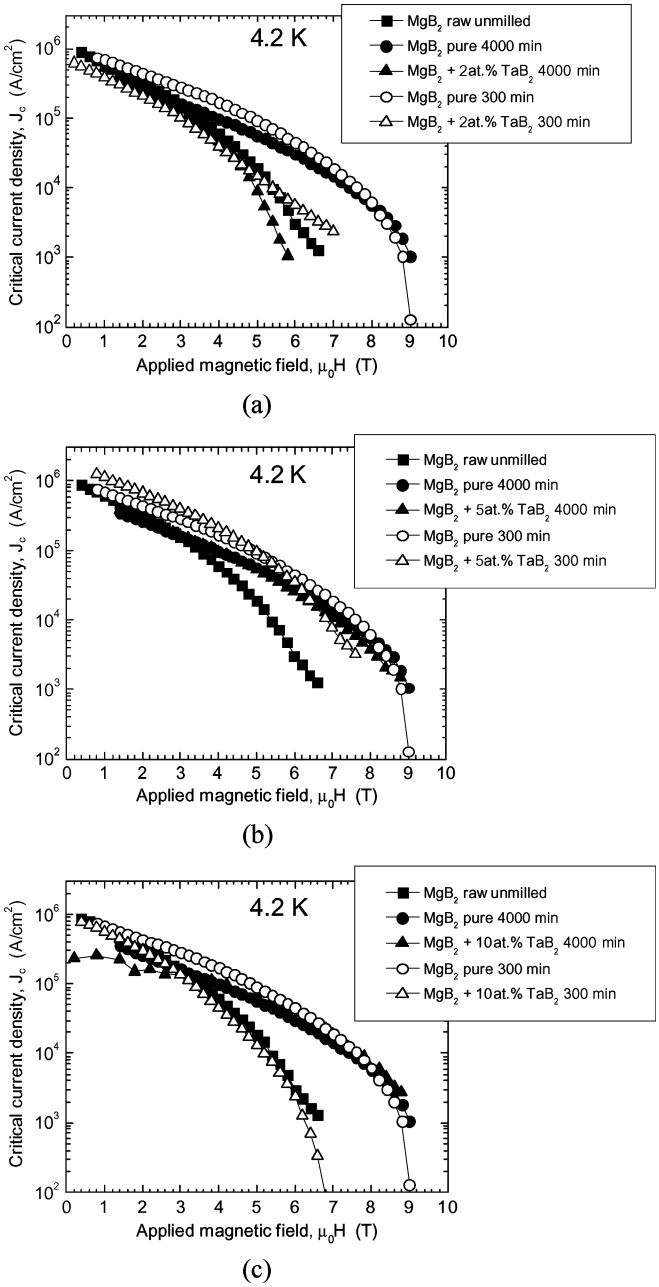

Fig. 1. Critical current density versus applied magnetic field for the unmilled and milled unmixed MgB_2 samples, and for the unmilled and milled TaB_2 mixed samples, all after HIPping. (a) $2\text{at.}\%TaB_2$, (b) $5\text{at.}\%TaB_2$, and (c) $10\text{at.}\%TaB_2$.

Fig. 2 shows the volumetric pinning force $F_p (= J_c B)$ as a function of applied magnetic field at 4.2 K for the HIPped samples. Fig. 2(a) shows samples mixed with $2\text{at.}\%TaB_2$. Mixing did not increase F_p above that for the raw material. It was found that samples that were only milled had higher F_p than the unmilled-unmixed and the mixed samples. These benefits are most probably due to milling-induced effects resulting in increased electron scattering. Milling can introduce defects in the superconducting phase, increasing its pinning force. Milling increased the maximum F_p from $\sim 6.5\ \text{GN/m}^3$, for the unmilled-unmixed sample, to $\sim 9\ \text{GN/m}^3$ for the unmixed sample milled for 300 minutes. Mixing decreased the pinning

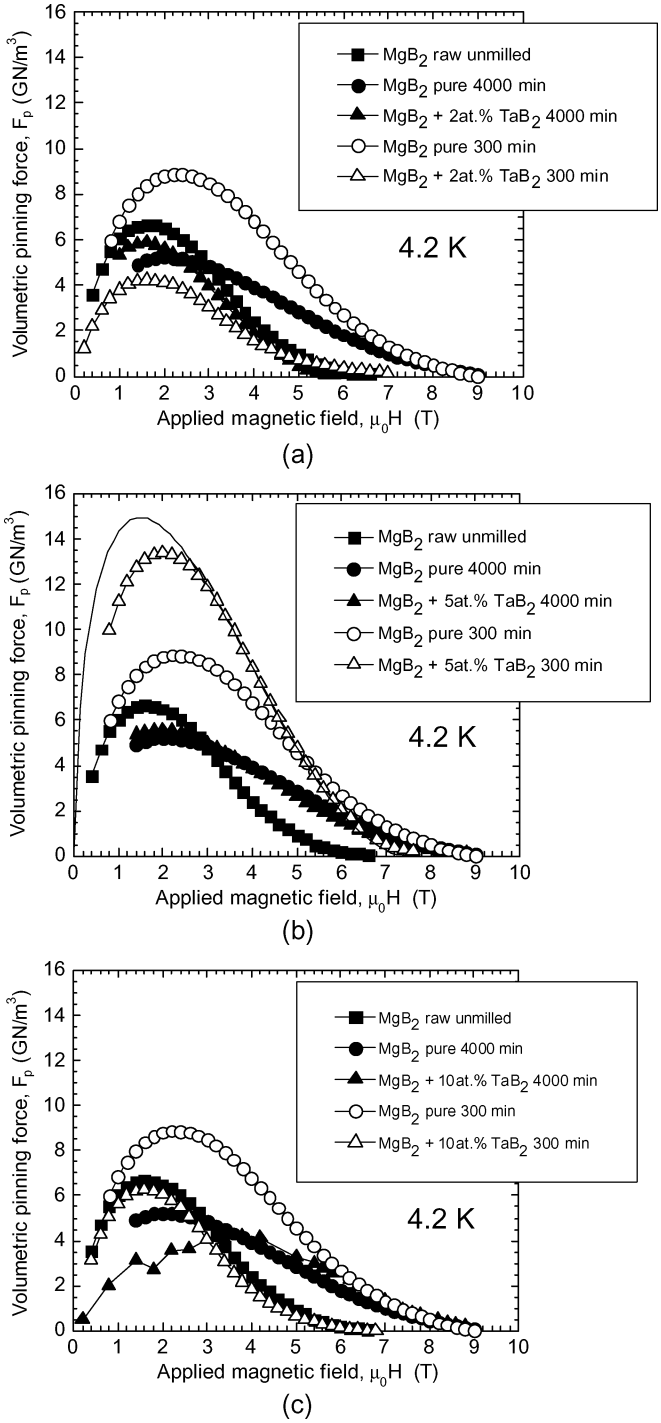

Fig. 2. Volumetric pinning force versus applied magnetic field for the unmilled and milled unmixed $\mathrm{MgB_2}$ samples, and for the unmilled and milled $\mathrm{TaB_2}$-added samples, all after HIPping. (a) $2\mathrm{at.\%\,TaB_2}$, (b) $5\mathrm{at.\%\,TaB_2}$, and (c) $10\mathrm{at.\%\,TaB_2}$. The solid line in (b) is explained in the text.

force compared to the unmilled-unmixed sample. The most important benefit from mixing and milling was the increase of $\mu_0 H_{irr}$, which increased J_c and F_p at high fields.

Fig. 2(b) shows samples mixed with $5\mathrm{at.\%TaB_2}$. In general, mixing and milling increased F_p for all samples in magnetic fields above 3 T. Milling effectively increased $\mu_0 H_{irr}$ through the increase of electron scattering and resistivity. Mixing with $5\mathrm{at\%TaB_2}$ and milling for short times (300 minutes) increased F_p at all magnetic fields, compared to the unmilled and to the unmixed samples. For all samples, milling was important to improve F_p and $\mu_0 H_{irr}$. The solid line (for the sample milled for 300 min) is a simulation of F_p as a function of $\mu_0 H$ using only the relation $F_p = K(T, B_{c2}) b^{1/2} (1 - b)^2$, where K is a constant that depends on T and B_{c2}, and $\mathrm{b} = \mathrm{B}/\mathrm{B_{c2}}$ is the reduced magnetic field. The approximation $B_{c2} = \mu_0 H_{irr}$ was used for these calculations. This simulation was performed to show the potential of the maximum pinning force supposing that grain boundaries are the only pinning centers. The poor agreement at low fields can be related to lower values of magnetization than those really expected for the samples.

This behavior is expected for samples whose pinning mechanism is mainly due to the presence of grains and the shearing of the flux line lattice on grain boundaries [13], [15]. There is good agreement for fields above the peak in the F_p curve for each sample. These results show that the pinning force has a peak close to 15 $\mathrm{GN/m^3}$ for the HIPped $5\mathrm{at.\%TaB_2}$ added sample milled for 300 minutes. As milling increased $\mu_0 H_{irr}$ by increasing the electron scattering and resistivity, the J_c vs. $\mu_0 H$ curves shifted to higher fields mainly because of the increase of $\mu_0 H_{irr}$, even though the pinning force was actually reduced in some cases. The reduction of the pinning force in these cases can be partially explained by the increase of the average particle size of the samples after milling and HIPping (Table I) compared to the unmilled HIPped sample. The milled samples could have larger grains after the same heat treatment mainly because of the increased internal energy of the milled $\mathrm{MgB_2}$ powder.

Fig. 2(c) shows samples mixed with $10\mathrm{at.\%TaB_2}$. Again, in general, mixing and milling increased F_p for all samples in magnetic fields above 3 T. Milling was effective to improve J_c and increase $\mu_0 H_{irr}$. However, mixing with $10\mathrm{at.\%TaB_2}$ and milling for short times did not improve the behavior in relation to the unmilled-unmixed sample.

The improvement of F_p for some of the $\mathrm{TaB_2}$-added samples could be related to the creation by milling of the $\mathrm{MgB_2}$ composite with finely disperse $\mathrm{TaB_2}$ particles, acting as efficient pinning centers and on the $\mathrm{MgB_2}$ grain refinement during HIP. This is also suggested by the almost constant $\mathrm{TaB_2}$ average particle size with milling.

The effect of the $\mathrm{TaB_2}$ additive, acting as doping or not, is not still well understood. As most of the improvements found in the mixed samples also can be related to the long milling times, the contamination by WC may be a source of reactive carbon in the ex-situ process, similarly to the SiC in the in-situ reaction [3]. The presence of carbon can induce boron substitution in the $\mathrm{MgB_2}$ lattice, improving $\mu_0 H_{irr}$ and J_c. This hypothesis is also corroborated by the reduction of the $\mathrm{MgB_2}$ a-parameter to around 3.078 Å after milling for 4000 minutes before HIPping, very similar to those found in C-doped $\mathrm{MgB_2}$.

IV. Conclusion

The use of high-energy ball milling to prepare $\mathrm{MgB_2}$ bulk samples is an useful method to study doping/addition and to optimize transport properties. High-temperature, high-pressure heat treatment (HIP) was used to produce high density samples with good connectivity.

Improvement of J_c at high magnetic fields could be obtained in milled samples mixed with TaB_2. Pinning was greatly improved for the milled and mixed samples. The sample added with $5\text{at.}\%TaB_2$ and milled for 300 minutes showed the highest values of J_c and F_p for fields below 6 T.

The milling increased electron scattering and resistivity, increasing $\mu_0 H_{irr}$ of the samples. The milled and mixed samples showed higher values of $\mu_0 H_{irr}$ than the unmilled and unmixed sample, even though several of the milled and mixed samples had lower pinning force as a result of their larger average grain size than the unmilled samples.

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