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Electrical and mechanical properties of superconducting MgB_2/Mg metal matrix composites

M Egilmez[1], L Ozyuzer[1,4], M Tanoglu[2], S Okur[1], O Kamer[3] and Y Oner[3]

[1] Department of Physics, Izmir Institute of Technology, Izmir, Turkey
[2] Department of Mechanical Engineering, Izmir Institute of Technology, Izmir, Turkey
[3] Department of Physics Engineering, Technical University of Istanbul, Istanbul, Turkey

E-mail: ozyuzer@iyte.edu.tr

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Abstract

MgB_2/Mg composites were prepared using a metal matrix composite fabrication method that offers the potential to produce superconducting wires as an alternative approach to the powder in tube process. To obtain composites, MgB_2 and Mg powders were mixed at different weight fractions and uniaxially pressed in a cylindrical die under the pressure of 0.5 and 1.0 GPa for two hours at various temperatures. The x-ray diffraction technique was used for phase identification. Temperature dependence of resistivity and magnetization measurements were carried out to determine superconducting properties. The effects of composite fabrication temperature and the addition of the Mg on the mechanical properties of MgB_2/Mg composites were investigated. For this purpose, the compressive mechanical testing was performed to measure the elastic modulus and fracture strength values of the composites. It was found that the relative weight fraction of the Mg and the fabrication conditions of the composites have considerable effect on the superconducting and mechanical properties of the composites.

1. Introduction

The discovery of superconductivity in MgB_2, which is a binary intermetallic compound, has offered a new superconducting material for practical applications [1]. The transition temperature, T_c, of MgB_2 (T_c = 39 K) provides a higher operating temperature as compared to currently available conventional superconductors such as Nb–Ti (T_c = 9 K) and Nb_3Sn (T_c = 18 K) for large scale applications [2]. However, MgB_2 exhibits low fracture toughness and hence brittleness as do Nb-based conventional superconductors and high temperature superconductors (HTS), and thus it is not an appropriate material for producing wires and tapes. The powder in tube (PIT) method has been a common commercial technique of producing wires and tapes from brittle superconductors. In this method, the brittle superconducting powder is filled into a ductile metal tube and then swaged into small diameters for various applications. This method has been widely used for both conventional superconductors and HTS. A number of research groups have recently reported successful fabrication of wires and tapes employing the PIT method with or without the application of a heat treatment [3–7]. Maintaining a critical temperature and critical current density (J_c) while improving mechanical properties of the superconductor during the mechanical deformation or following the annealing process in PIT is essential. Occurrence of any reaction between the metal and superconducting phases may affect the superconducting properties of the wires. For this reason, analysis of substitutional chemistry of the superconducting material is crucial. Accordingly, the PIT method has been used to produce MgB_2/metal composite wires with various metal sheaths such as stainless steel [6], Cu [4], Ag [4], Cu–Ni [6] and Fe [8, 9]. Iron was determined to be the best sheath material for MgB_2 wires, providing high critical current density of 1.42×10^5 A cm^{-2} (4.2 K and 4 T) and inertness to MgB_2 even with annealing at 900°C [10].

[4] Author to whom any correspondence should be addressed.

An alternative approach to the PIT method is the utilization of the metal matrix composite fabrication methods. Metal matrix composites, in general, consist of at least two components; i.e. one obviously is the metal matrix, and the second component is in general a ceramic or an intermetallic compound in the particulate, whisker or short fibre forms. The metal matrix maintains the form of the composite, acts as a load carrying medium and improves ductility and the fracture toughness of the composite while the particulate constituent enhances the properties such as the wear resistance, hardness and elastic modulus values. Although metal matrix composite processing routes are not a widespread routine for producing bulk superconductors, there are a few studies reported using this method for HTS in Ag metal matrix [11] and MgB_2 in Al and Mg metal matrices [12, 13]. However, to our knowledge there is no work in the literature investigating the mechanical behaviour of the brittle superconductor composites. In this study, MgB_2/Mg metal matrix composites were prepared by blending superconducting MgB_2 with Mg, that provides ductility and low melting point. Electrical, microstructural and mechanical properties of the composites prepared under various conditions were characterized.

2. Experimental details

Commercial MgB_2 (Alfa Aesar) and Mg powders (−270 mesh and 99.9% purity) as matrix materials were used for the fabrication of MgB_2/Mg composites. The Mg amount in the composites was selected as 0, 5, 10, 15 and 20 wt% for different sets of samples. MgB_2 and Mg powders were mixed homogenously and uniaxially pressed under the pressure of 0.5 and 1.0 GPa in a steel die. The die was heated to 400 and 500 °C and kept at these temperatures for 2 h under pressure in air. It was revealed in the literature that although the oxidation of MgB_2 in air begins at 400 °C, it is not very extensive up to 700 °C [14]. Therefore, the selected temperatures for producing MgB_2/Mg composites were expected to be suited to prevent the oxidation of MgB_2 and Mg to MgO. Since the melting point of Mg is 654 °C, no preponderant oxidation reaction was expected for Mg at 400 and 500 °C. During the preparation of pellets, those with the thickness of 1 mm and the diameter of 15.6 mm were prepared for electrical characterizations. In addition, pellets with the thickness of 7 mm and the diameter of 10 mm were fabricated for mechanical testing. The x-ray diffraction (XRD) technique was used for phase identifications. For this purpose Philips™ XRD equipment was operated using Cu Kα radiation. Resistivity versus temperature measurements were performed using the rectangular pieces (15.6 × 3.0 × 0.9 mm^3 in size) of the samples cut from the pellets. The resistivity measurements were carried out in a closed cycle He refrigerator. Magnetization measurements were made using a vibrating sample magnetometer under 180 G and magnetic field cooling. Mechanical tests were performed using a Shimadzu™ universal mechanical test machine on samples prepared under 0.5 GPa at 400 and 500 °C. The fracture strength and compressive elastic modulus values were determined from the stress versus strain curves of the composites.

Table 1. Density of the MgB_2/Mg composites.

Pressure (GPa)	Temp. (°C)	Mg (%)	Density (g cm^{-3})
0.5	400	0	1.85
0.5	400	5	1.86
0.5	400	10	1.90
0.5	400	15	1.93
0.5	400	20	1.96
0.5	500	0	1.85
0.5	500	5	1.86
0.5	500	10	1.91
0.5	500	15	1.92
0.5	500	20	2.01
1.0	400	0	2.10
1.0	400	5	2.10
1.0	400	10	2.11
1.0	400	15	2.10
1.0	400	20	2.12
1.0	500	0	2.15
1.0	500	5	2.17
1.0	500	10	2.10
1.0	500	15	2.17
1.0	500	20	2.12

3. Results and discussions

The densities of the prepared MgB_2/Mg composites are given in table 1. The theoretical densities of MgB_2 and Mg are 2.62 and 1.74 g cm^{-3}, respectively. Although the addition of low density Mg is expected to reduce the density of the composites, the measured values indicate a contradictory result. As the Mg amount is increased, an increase of the density is measured due to the reduction of porosity in the structure. During pressing at elevated temperatures, Mg metal flows and fills the pores that exist within MgB_2 compacts. Similarly, the increase of temperature and pressure improves the density values by reducing the porosity level.

The powder XRD patterns of the MgB_2/Mg composites prepared with various concentrations of Mg at 500 °C under the pressure of 0.5 and 1.0 GPa are shown in figures 1(a) and (b), respectively. The intensity of each pattern is normalized with respect to their maximum. In the same graphs, the patterns of Mg and commercial MgB_2 powders are also presented for comparison. As seen in the figures, the main phase for MgB_2/Mg composites is MgB_2. As the amount of Mg increases in the composites, Mg peaks become detectable in the spectrum as seen in the pattern. It can be noticed that there is no major reaction between MgB_2 and Mg. By comparing the commercial MgB_2 powder with the MgB_2 after pressing, it was found that no detectable oxidation reaction develops during pellet preparation stages. However, the presence of a small amount of MgO secondary phase is seen in the pattern of Mg substituted composites. MgO might arise during the heat treatment of MgB_2 and Mg mixture. This minor amount of MgO is expected to enhance the superconducting properties of the composite wires acting as pinning centres and increase the critical current density, J_c. Also, the formation of MgB_4 secondary phase in the case of hot pressing was revealed in the literature [14]. The existence of MgB_4 in superconducting composite is undesirable, because it is generally situated around grain boundaries as an insulating layer and degrades the superconducting properties. The MgB_2/Mg composites prepared with various concentrations of Mg at 400 °C under the

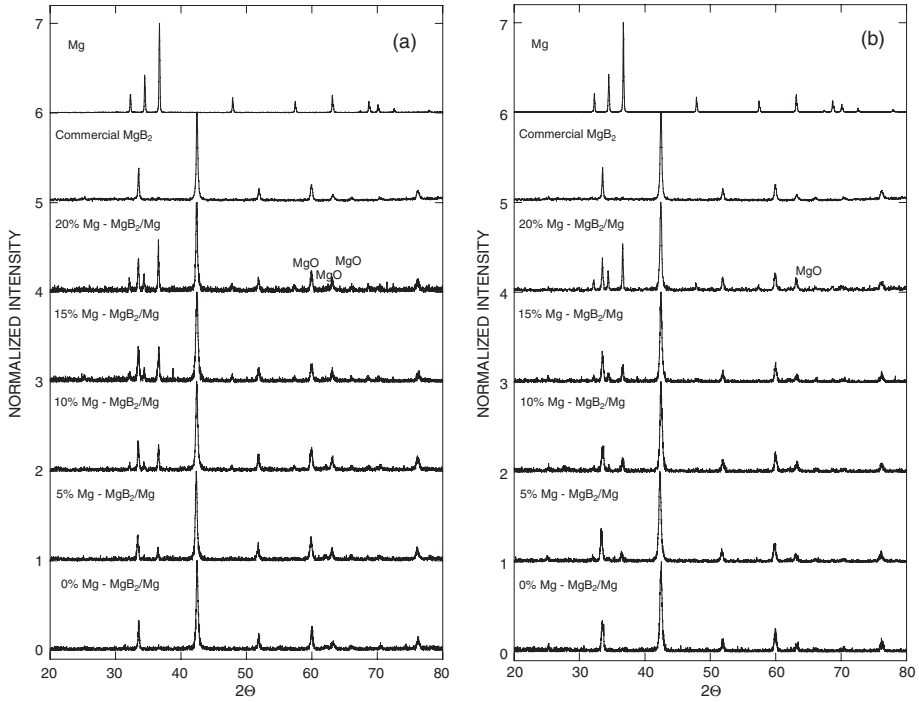

Figure 1. ((a), (b)) Powder XRD pattern of Mg, commercial MgB_2 powder and MgB_2/Mg composites prepared with different Mg ratios prepared at 500 °C and under pressures of (a) 0.5 GPa and (b) 1.0 GPa.

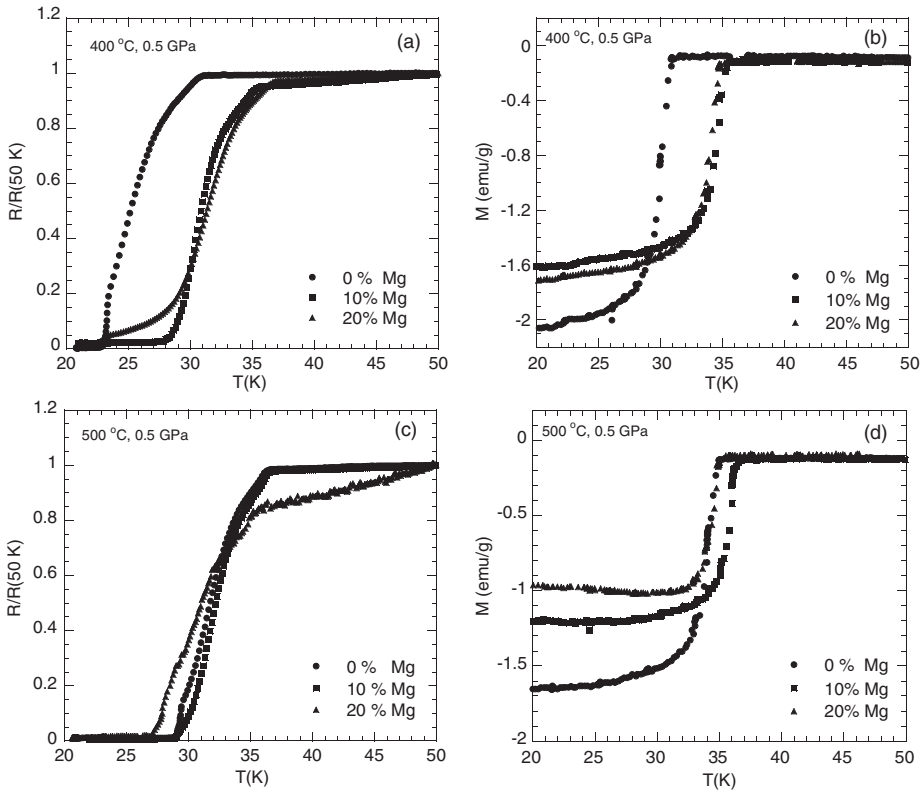

Figure 2. ((a)–(d)) Resistivity–temperature and magnetization–temperature curves of MgB_2/Mg composites prepared at 0.5 GPa.

pressures of 0.5 and 1.0 GPa are not presented here; however, they exhibited similar XRD patterns as for 500 °C.

It is vitally important to maintain superconducting properties of the MgB_2 in the case that it is fabricated as a metal matrix composite wire. In the present study, we investigated the resistivity and magnetization of the composites in order to compare with pure MgB_2. Figures 2(a)–(d) show the resistivity versus temperature and magnetization versus

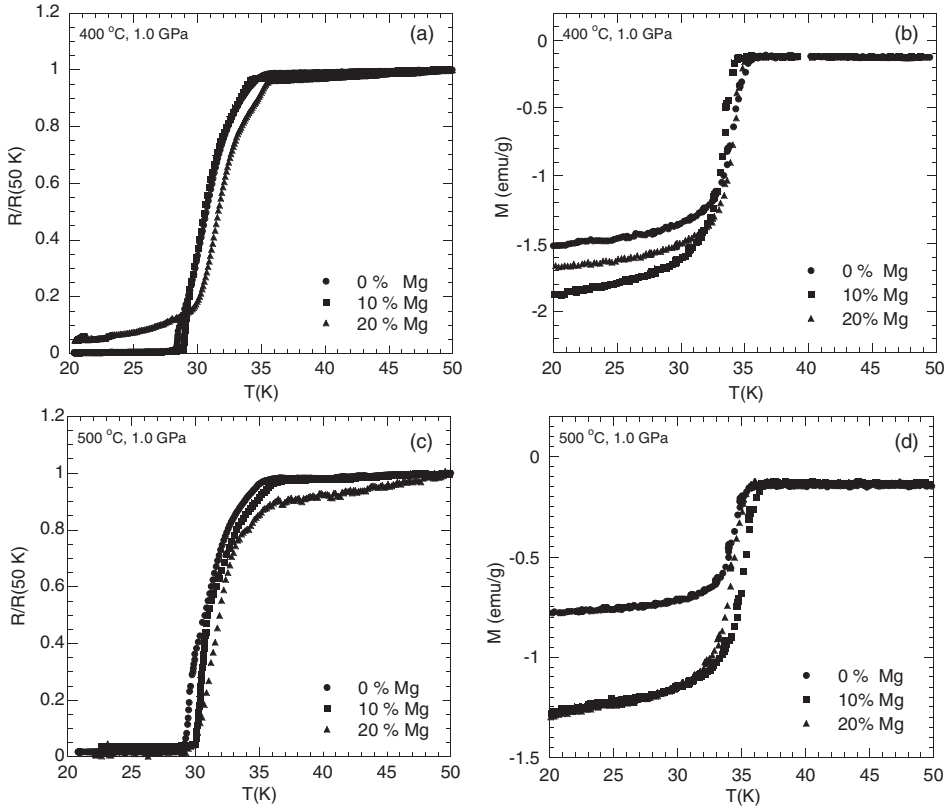

Figure 3. ((a)–(d)) Resistivity–temperature and magnetization–temperature curves of MgB_2/Mg composites prepared at 1.0 GPa.

temperature curves of the samples prepared under 0.5 GPa pressure at 400 and 500 °C. The resistivity values in these figures are normalized to their values at 50 K. In the figures, the resistivity and magnetization results are not presented for 5% and 15% Mg addition for clarity since they show similar trends. As seen in figure 2, the Mg amount in composites decreases the slope of the resistivity curve above the transition temperature, since the resistivity of Mg has a strong dependence on temperature. Also, Mg has lower resistance (1.6 $\mu\Omega$ cm at 300 K) as compared to MgB_2 (4.5 $\mu\Omega$ cm at 300 K). Therefore, the increasing fraction of Mg decreases the resistivity of MgB_2/Mg composites.

Figure 2(a) shows a T_c^{Onset} value of 30.9 K for the sample without Mg. It is observed that increasing the Mg content increases the critical temperatures of the composites. It was revealed that all of the Mg substituted samples have critical temperatures greater than the unsubstituted sample. The highest T_c^{Onset} (35.6 K) was obtained from the sample prepared at 400 °C under pressure of 0.5 GPa with 10 wt% excess Mg. Thus, the improvement in the T_c values due to decrease in the porosity ratio is expected to continue until a certain amount of substitution. Moreover, the composites presented in figure 2(a) have large superconducting transition width (ΔT values, temperature change in 10%–90% resistivity drop) varying between 4.6 and 6.2 K. From figure 2(c), it is seen that the unsubstituted sample has a T_c^{Onset} value of 36.1 K. Similar to the samples prepared at 400 °C under pressure of 0.5 GPa, the highest T_c^{Onset} value (36.8 K) was observed in samples prepared at 500 °C under pressure of 0.5 GPa with 10 wt% excess Mg. However, increasing the amount of substituted Mg decreases the volume fraction of the superconducting phase within the composites, which is obtained from magnetization measurements given in figures 2(b) and (d). As can be seen in table 1, an increase of 100 °C for the sample pressing temperature under the pressure of 0.5 GPa results in no major effect on the density of the samples. However, this much increase of temperature resulted in an increase of 5.2 K in T_c^{Onset} value.

We would like to emphasize that the variation of T_c^{Onset} values was found to be not so large for the substituted samples as in unsubstituted samples. Figure 3(a) demonstrates the temperature dependence of resistivity of the samples prepared at 400 °C and 1.0 GPa. A relatively sharp transition is seen, if it is compared with the samples prepared at 0.5 GPa. The ΔT value was found to be improved by 1.4 K for unsubstituted sample for composites prepared at 1.0 GPa as compared with the composite prepared at 0.5 GPa for 400 °C pressing temperature. Considering the SEM results which were reported earlier [15], it can be realized that increasing pressing temperature increases grain connectivity between MgB_2 grains, therefore a less broad transition is observed. For the samples prepared at 400 °C and 1.0 GPa, it is observed that for all the samples the T_c^{Onset} temperature is very close to or higher than 35 K. These results are better than samples prepared with 0.5 GPa pressure. The T_c^{Onset} was observed in the sample with 5 wt% excess Mg for 400 °C and 1.0 GPa samples with 37.4 K. Similar to 400 °C and 1.0 GPa samples, the best T_c^{Onset} value (37.3 K) was observed for the sample with 5 wt% excess Mg for 500 °C and 1.0 GPa. For all samples presented in figure 3(c), T_c^{Onset} values are above 35 K, and T_c values are

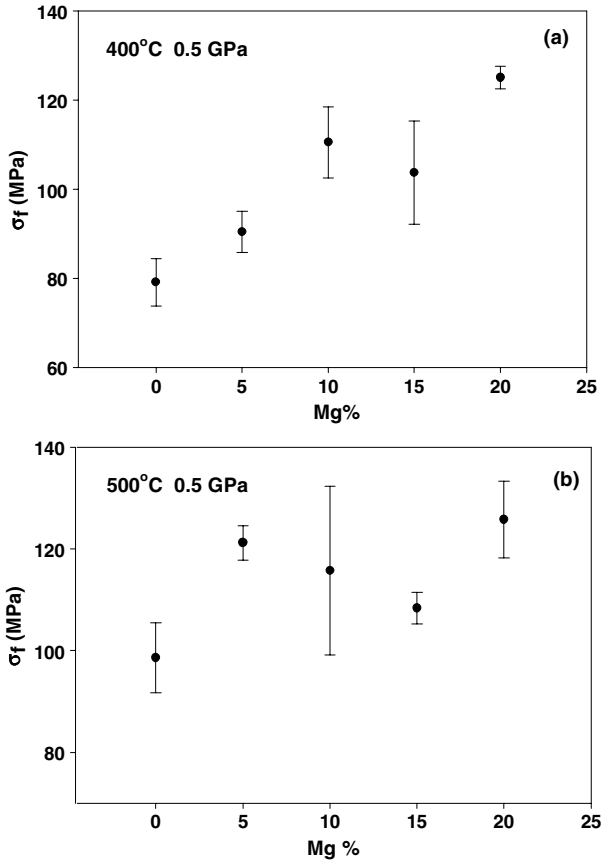

Figure 4. Apparent strength versus Mg% for MgB_2/Mg composites prepared under 0.5 GPa pressure, at (a) 400 °C and (b) 500 °C, respectively.

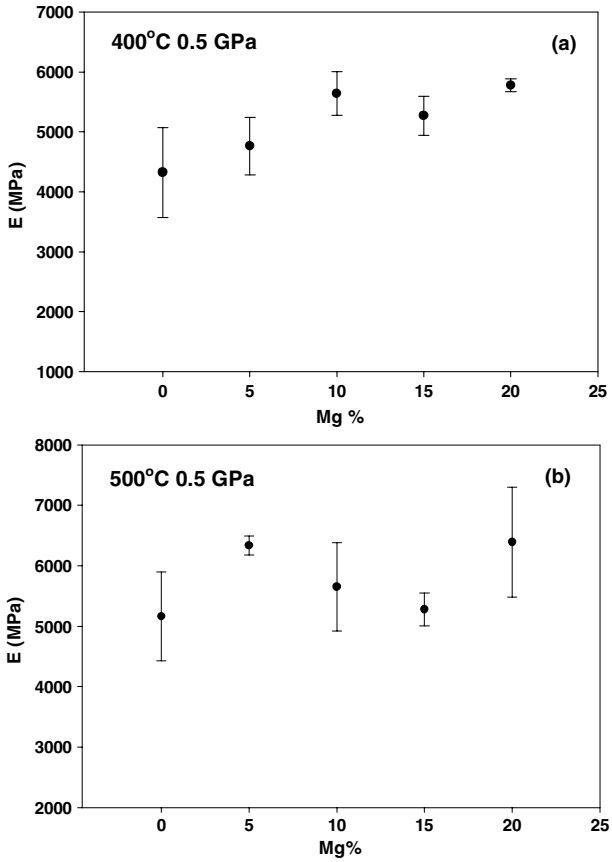

Figure 5. Elastic modulus versus Mg% for MgB_2/Mg composites prepared under 0.5 GPa pressure, at (a) 400 °C and (b) 500 °C, respectively.

Table 2. Resistivity of the MgB_2/Mg composites at 50 and 300 K.

Pressure (GPa)	Temp. (°C)	ρ (50 K) (mΩ cm)	ρ (300 K) (mΩ cm)
0.5	400	1.57	1.91
0.5	500	1.60	1.88
1.0	400	1.24	1.47
1.0	500	1.03	1.25

above 30 K even in the sample with 20 wt% Mg. This means that we are well below the percolation threshold of MgB_2/Mg composites. A critical temperature T_c above 30 K is significant for large-scale applications.

The resistivity of the unsubstituted samples for all sets of MgB_2/Mg composites is given in table 2. Recently, the resistivity behaviour of MgB_2 and their temperature dependences for thin films, single crystals and polycrystalline samples have been reviewed in the literature [16]. A wide range of resistivity values, between a few $\mu\Omega$ cm and 100 mΩ cm, were reported for bulk samples. The spread of measured resistivity may be associated with the porosity, structural defects and preparation conditions. Since the resistivities for our samples are in the range of mΩ cm, the high porosity ratio of the samples may be related to the measured values.

The mechanical behaviour of the brittle MgB_2 is critical for various applications, especially superconducting wires and tapes. To obtain MgB_2 based superconducting systems with improved fracture toughness, Mg was blended with MgB_2 to prepare composites. The compressive mechanical testing was performed to obtain elastic modulus (E) and fracture strength (σ_f) values of the MgB_2/Mg composites. The variation of σ_f and E values of the composites as a function of increasing Mg amount are given in figures 4 and 5, respectively. It was found that, in general, both σ_f and E values increase as the Mg addition is increased in the composites. Similarly, it was observed that the mechanical properties are improved as the pressing temperature during composite preparation is increased (from 400 to 500 °C). As an example, average apparent σ_f values of 80 and 128 MPa were measured for the composites prepared without and with 20 wt% Mg addition, respectively, at pressing conditions of 400 °C under 0.5 GPa. For these conditions, the σ_f values increase by up to 56% with the addition of 20 wt% Mg to MgB_2, as compared to those prepared without Mg addition. Similarly, an improvement of the E values was found as the Mg content was increased. The average E values increased by 38% as 20 wt% Mg is blended in (4.5 and 5.8 GPa for the composites without and with 20 wt% Mg addition, respectively). Moreover, the strain of failure (ε_f) values were measured as 0.025 ± 0.001 and 0.035 ± 0.005 for the composites prepared without and with 20 wt% Mg addition. The improvement of σ_f, E and ε_f values is associated with reduction of the porosity within the composite structure

and a better compaction of the MgB_2 powders due to the presence of the Mg metal. During the hot pressing process, Mg flows around MgB_2 powders and forms a matrix that holds the powders together and provides load transferring medium. It was also measured that the pressing temperature has some effects on the mechanical behaviour of the materials. As seen in the figures, as the pressing temperature increases, σ_f, E and ε_f values, in general, increase slightly with some fluctuations. Further increase of the Mg content may be expected to obtain composites with better ductility; however, the percolation threshold of MgB_2/Mg composites restrict us to blend in a higher amount of Mg. Superconductivity of the composites is expected to degrade significantly with the addition of Mg above 20 wt%.

Hot isostatic pressing was used by several groups to prepare dense MgB_2 samples [17]. In their studies an elastic modulus value of 79 GPa was reported for the samples prepared with a hot isostatic press and a density of 2.39 g cm^{-3}. Although there is a limited number of studies on the mechanical properties of PIT wires, an elastic modulus value of 29 GPa was reported for Fe clad MgB_2 wires [18]. In our samples, although a densification behaviour was observed by increasing the amount of additional Mg, the measured densities (between 1.80 and 1.88 g cm^{-3}) and the elastic modulus values (between 4 and 6 GPa) are still lower than what is expected.

4. Summary

It was found that metal matrix composite fabrication routes have great potential to prepare MgB_2/Mg composites. The pores within the MgB_2 powder compacts are filled by the flow of Mg under pressure of elevated temperatures. This results in an increase of the density of the composites. XRD results indicate that no major reaction between MgB_2 and Mg occurs under the conditions applied within the study. The superconducting properties of MgB_2/Mg metal matrix composites have been investigated with resistivity versus temperature and magnetization versus temperature measurements. It has been found that addition of Mg in MgB_2/Mg metal matrix composite improves the T_c and normal state properties of samples. However, a higher amount of Mg, which is substituted up to 20 wt%, decreases the superconducting fraction of the composites. Since the mechanical properties of the brittle MgB_2 are critical for industrial applications, the results revealed that blending the MgB_2 with Mg provides ductility to the system. In general, fracture strength, elastic modulus and strain at failure values increase with the incorporation of Mg due to the reduction of the porosity and the enhancement of the compaction of the powders.

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