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Development of Magnesium Diboride Superconducting Wires through Hot Working with Different Initial Filling Density

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Abstract. This study aimed to investigate the use of the hot working in a sealed tube method for the production of Magnesium Diboride (MgB_2) wires from a powder state. The wires were synthesized using different initial filling densities of 60%, 80%, and 100%. The Magnesium and Boron powder was ground using agate mortar into a stoichiometric mixture by weight of Mg:B=1:2 and then packed into a stainless steel (SUS316L) tube. Subsequently, the pack was sintered for two hours at 800°C in an air atmosphere and continuously rolled to form a wire. XRD and SEM analyses were then conducted to observe the phase development of the sample produced. The diffraction pattern and microstructure observation results showed that MgB_2 phase was successfully created using economically advantageous raw materials of crystalline Mg and amorphous B with 60% filling density. The size of the crystallites and superconducting phase was shown to experience a significant increase. The R-T cryogenic magnet assessed the sample of critical temperature, and MgB_2 produced using full amorphous boron had a transition temperature of 39 K.

Keywords: Critical temperature; Filling density; Hot working; MgB_2 Superconductors; Wire

1. Introduction

Magnesium diboride, MgB_2, is a novel high critical temperature superconductor with a critical temperature of 39 K, initially introduced in the early twenty-first century (Nagamatsu *et al.*, 2001). The versatility of MgB2 also extends to its production in various forms and shapes, including bulk (Arvapalli *et al.*, 2021; Zhang *et al.*, 2020; Zheng *et al.*, 2019), thin films, tapes, and wires (Balog *et al.*, 2018; Grivel, 2018b; Herbirowo *et al.*, 2018; Kováč *et al.*, 2011; Vinod *et al.*, 2009). Consequently, this material has found promising applications in various electronic devices due to its excellent electrical contact between grains and cost-effectiveness. Several studies have shown that its superconductivity has a between grains and cost-effectiveness. Several studies have shown that its

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superconductivity has a higher critical temperature compared to niobium-based superconductors, such as Nb_3Sn and NbTi. This makes MgB_2 a potential substitute in the production of superconducting devices, such as Magnetic Resonance Imaging (MRI). Previous studies has proposed a direct method for crafting superconductor wire or tape through the Powder-in-tube (PIT) method (Kováč *et al.,* 2020; Flukiger, Hossain and Senatore, 2009). This method can be implemented either in situ, comprising the filling of a metal tube with a mixture of Mg and B. The process is then continued with isolative sealing and mechanical deformation to release the mixture materials from the air. The PIT method can also be achieved ex-situ, where MgB_2 powder is filled into the tube and rolled or drawn. According to Mustafa Akdogan et.al, the critical current increases along with the infill density (Akdogan *et al.,* 2015). Gadja et al. also stated that the initial filling density of MgB2 could increase the current density (Jc) and the connectivity between particles (Gajda *et al.,* 2021). Therefore, this study presents recent results on the fabrication of MgB_2 wire using the PIT method and hot working deformation through hot rolling. The main objective is to synthesize a material with a great microstructure and phase composition that maximizes superconductivity. The results of a cryogenic magnet system developed by Oxford Instruments showed the critical temperature of MgB_2 wire, suggesting its potential MRI application.

2. Methods

The powder-in-tube (PIT) technique was used to create MgB_2 monofilament wire (Lubis *et al.,* 2018; Glowacki *et al.,* 2001). The feedstocks used included magnesium powder (Sigma Aldrich, 98% purity) and amorphous low-cost boron powder (Luoyang China, 95% purity). Furthermore, the SUS316L tube with an outer diameter and inner diameter of 6 and 4 mm, respectively were used as a container for MgB_2 powder with a ratio Mg:B = 1:2. The powders were then mixed and ground in an agate mortar for 30 minutes in an environment of air. The SUS316L stainless steel rod (4 mm in diameter) was used to seal the stainless-steel tube container after it had been filled with the mixture. The SUS316L tubes in wire product manufacturing offered the advantage of enhanced corrosion resistance and durability, ensuring a longer lifespan and reduced maintenance requirements for the final product (Widyianto, Baskoro and Kiswanto, 2022; Anwar *et al.,* 2021). The coarse-grained SUS 304 had a higher surface roughness ratio compared to the coarse-grained SUS 316. The fine-grained SUS 304 and the fine-grained SUS 316 had comparable inhomogeneous grain strengths (Abdul *et al.,* 2021). Hot rolling was used to create 3 mm square rods, introducing a novel method that enabled the formation of the desired rod geometry but also effectively mitigated strain hardening, thereby streamlining the overall manufacturing process. Hot working in superconducting wire manufacturing was crucial for achieving densification, consolidating the powder, refining the grain structure, improving mechanical properties, and shaping the wire into the desired form. A wire specimen was sintered for one hour at 1073 K in a muffle furnace. The hot rolling process in this study showed that the wire that was still hot from the furnace was taken out directly to be rolled by the rolling machine. X-ray diffraction (Smartlab Rigaku) was used to determine the phase of materials. The Field Emission Scanning Electron Microscope - EDX (JEOL JIB 4610F) was used to examine the specimen's morphology, porosity, and elemental composition. The four-point probes method and Oxford Instrument Teslatron PT's cryogenic magnet apparatus were used to measure temperature dependence resistivity. Furthermore, the device was used to measure resistivity at temperatures between 8 and 200 K for the samples.

3. Results and Discussion

X-ray diffraction was used to identify MgB_2 wire that had been sintered at 800°C and then hot rolled. The wire was cut transversely to obtain MgB_2 from inside the wire for XRD measurement. Figure 1 depicted the metallography preparation of the SUS316L tube wire, followed by the diffraction pattern of MgB_2 powder. The pattern showed that MgB_2 phase was one of the main phases, which increased from 98.2% to 98.5% and contained other elements, with a small quantity of Fe impurities. This proved that the SUS316L sheath was in contact with the diffraction process. According to Varghese et al., unreacted Fe and SS in MBFe and MBSS provided a greater volume percentage of MgB_2 core, superior current density characteristics, and flux pinning behavior. This showed that Fe and SS were better sheath materials for the manufacture of MgB_2 wire and tape (Varghese *et al.*, 2007).

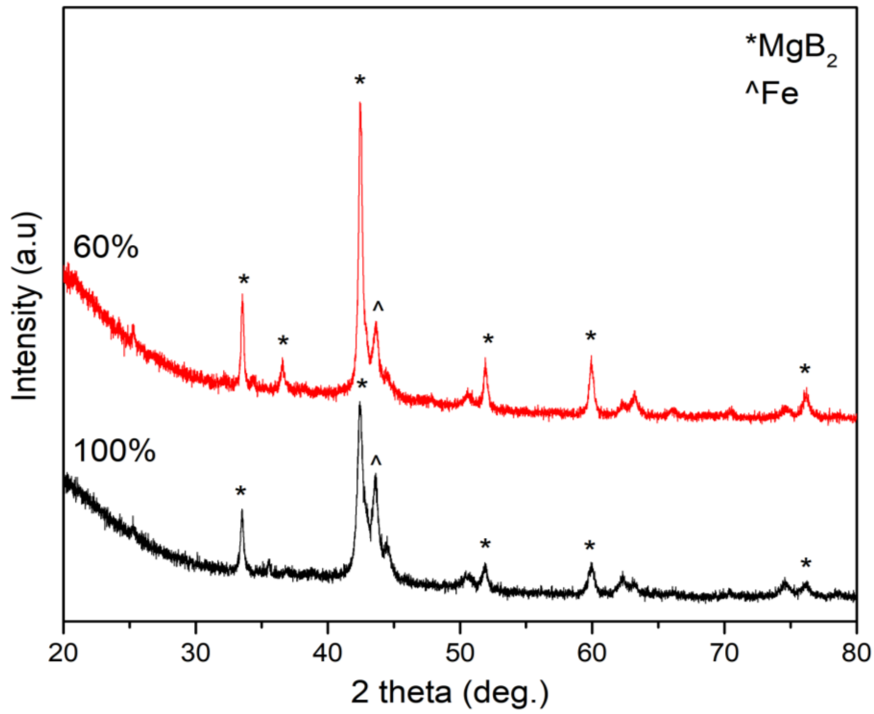


Figure 1 The diffraction pattern and difference plot of MgB_2 wire with 100% and 60% initial filling density

The largest current density was obtained when filling density was increased from 50% to 60% of the theoretical mass density of Mg+B in a previous study (Akdogan *et al.*, 2015). However, the filling mass density had a significant impact on the temperature of production. Based on these results, the ideal filling density for powder was less than 100%. To prevent the porosity or size reduction of MgB_2 wire, MgB_2 formation problem must be addressed during sintering.

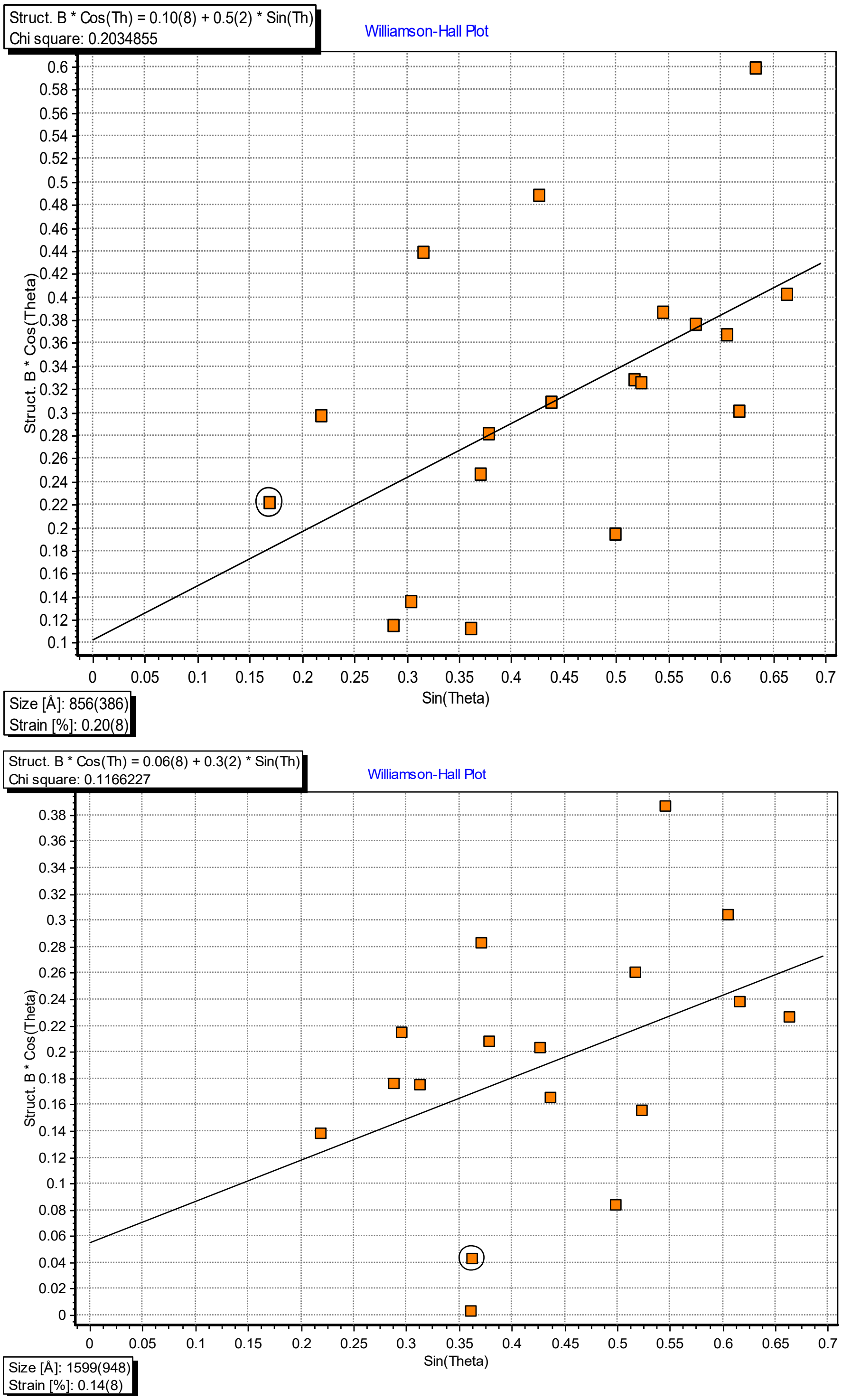


Figure 2 FWHM plot for sample MgB_2 wire with 100% (upper) and 60% (lower) filling density

Figure 2 showed the W-H plot of MgB_2 superconducting wire with various variations of powder filling, namely 100% and 60%. Analysis of the Williamson-Hall method for this computational analysis gave an increase in crystallite size from 85.6 nm to 159.9 nm, with a slight decrease in the amount of crystallite strain. This showed that slightly higher oxide formation tended to increase the crystallinity of the wire towards the ceramic material (Herbirowo *et al.*, 2023). MgB_2 wire specimen for morphological analysis is presented in Figures 3 and 4. Furthermore, the results showed that the porosity decreased from 6.3% to

5.4%, as shown in Figure 4. Based on Figure 3, it was easy to detect the location of the SUS316L tube and MgB_2 phase boundaries. The results showed that the SUS316L did not react with Mg and B precursors. The cross-section line scan with EDX shown in Figure 3 also showed the presence of oxide in the material. Compared to XRD results, which did not show any oxide phase, it was estimated that the oxygen element in the EDS results was due to the oxidation process occurring on the surface of MgB_2 sample.

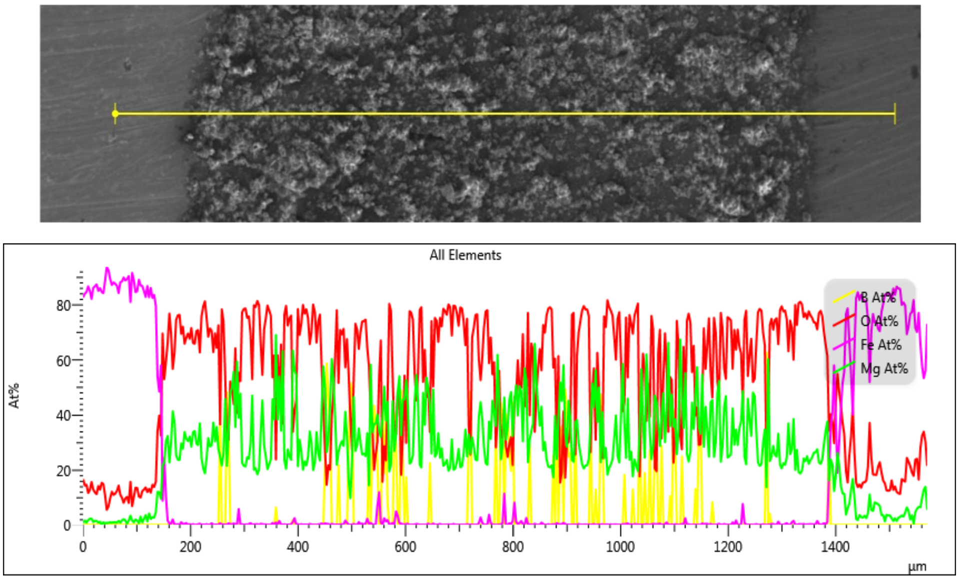
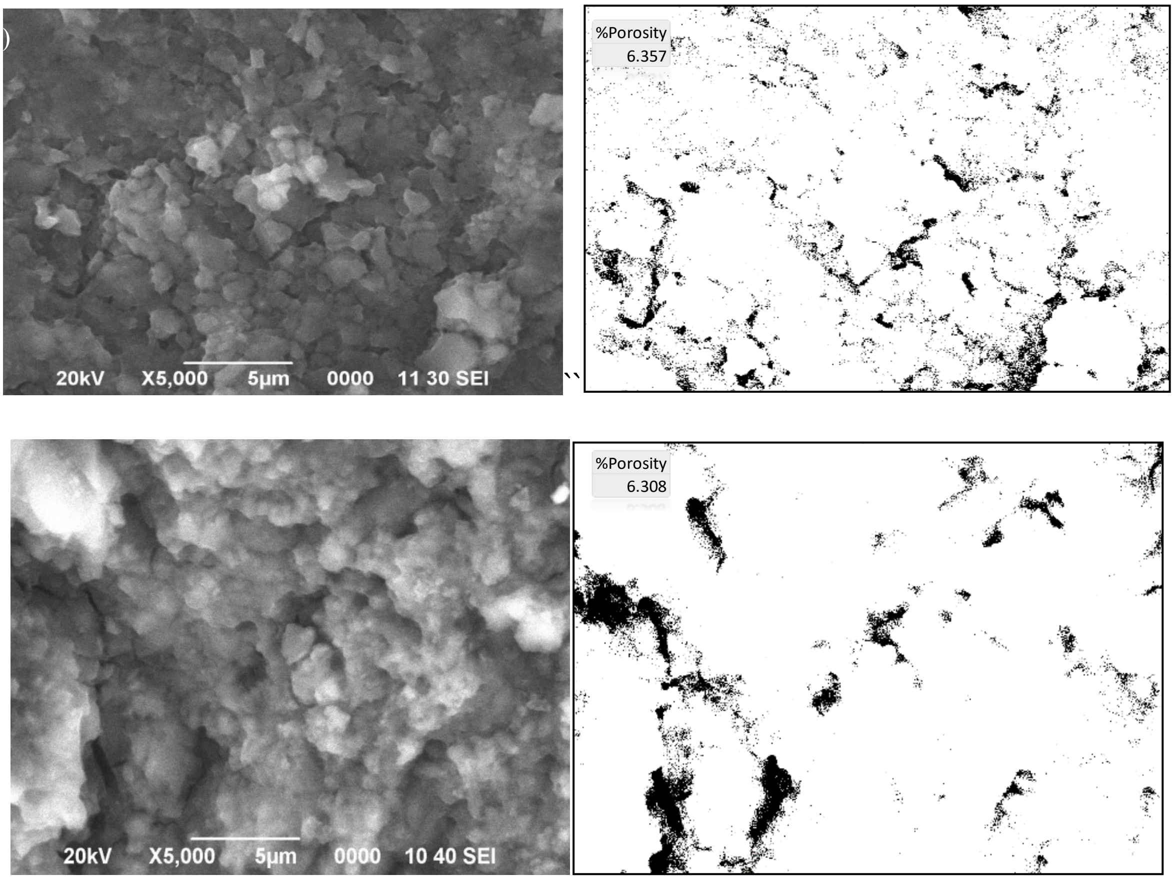


Figure 3 EDX line scan on cross-sectional MgB_2 wire of 60% filling density with a magnification of 20 times



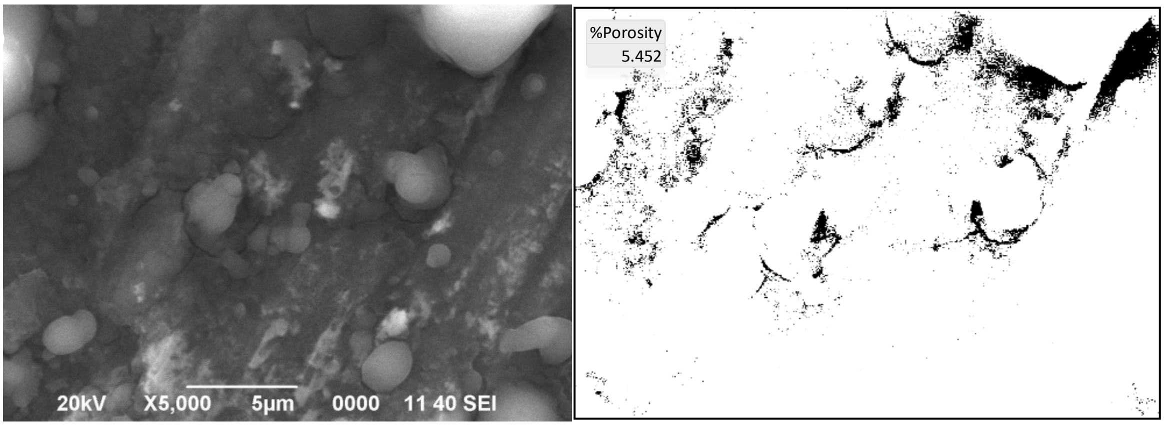


Figure 4 The microstructure of the SUS316L/MgB_2 wire sintered at 1073K for 1 hour through hot rolling with (a) 100% (b) 80%, and (c) 60% filling density under a magnification of 5.000 times on the left (Secondary Image) and right (porosity analysis) sides

Table 1 The objective of MgB_2 synthesis was to create a material with a microstructure, phase, and composition with the highest levels of superconductivity. The electrical resistivity's temperature dependency was one of the most important properties to be assessed. Furthermore, all synthesized MgB_2-based samples had their resistivity evaluated using a cryogenic magnet setup and the four-point probe (FPP) method. The electrical resistivity of the SUS316L/MgB_2 monofilament wire was shown as a function of temperature in Figure 5. The results showed that the resistance value of the SUS316L/MgB_2 wire sample with 60% filling density was initially 2.00×10^{-4} ohm at 200 K and decreased continuously until 8 K to 1.50×10^{-4} ohm, before dropping at Tc_{onset} of 39K.

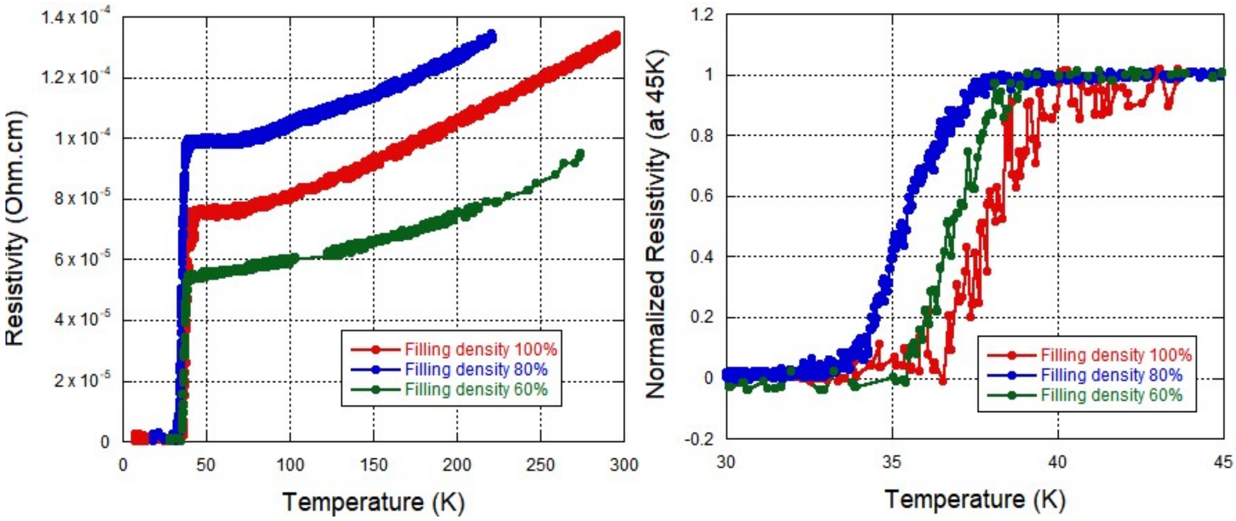


Figure 5 Resistivity measurement (left) and normalized resistivity (right) of the SUS316L/MgB_2 wire specimen with different initial filling densities without an applied magnetic field

4. Conclusions

In conclusion, MgB_2 wires were fabricated successfully, leveraging the economic benefits of using raw materials from crystalline Mg and amorphous B, with a 60% initial filling density. Furthermore, the size of the crystallites and superconducting phase greatly increased in this study. The R-T cryogenic magnet assessed the specimen of critical temperature, and the results showed that MgB_2 specimen created using crystalline magnesium and fully amorphous boron had a transition temperature of 39 K.

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