MANUFACTURING AND DEVELOPMENT OF MgB_2 SUPERCONDUCTING WIRE THROUGH HOT ROLLING PROCESS

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ABSTRACT

In this study, the synthesis and manufacture of MgB_2/SS 316L Superconducting Wire by In-Situ method through hot rolling with a variety of sintering temperatures and holding time with the novelty of the SS316 sheath and hot rolling method were carried out to maintain the size reduction results without stress concentration and minimal defects. MgB_2 is a superconducting material with a critical temperature of ~39 K. Hot rolling process with a variety of sintering temperatures and holding time was carried out to observe its effect on the superconductivity characteristics of MgB2. The process of synthesis and sample preparation begins by measuring and cutting the SS3I6L tube and weighing the raw materials, which were Mg and B powders with a ratio of 1:2 according to stoichiometric calculations. The material was then grounded for 2 hours using an agate mortar and then put into an SS 316L stainless steel tube where it was completely compacted. Then, the sintering process was carried out using a muffle furnace with variations in sintering temperature of 780°C, 800°C, 820°C, and holding time for 1 hour and 3 hours. After that, a hot rolling process was carried out on each sample with a temperature of 300°C. The samples were characterized by XRD, Cryogenic Magnet and SEM-EDS. The identification through XRD showed that the phases formed were MgB2, MgO, Fe. The Cryogenic Magnet test showed superconductivity at a sintered temperature of 800°C and 820°C with a holding time of 1 hour, where for a sintered temperature of 800°C, it has a Tc^{onset} of 39.55 K and a Tc^{zero} of 34.69 K while at sintered temperature of 820°C the Tc^{Onset} is 38, 44 K Tc^{Zero} is 31.30 K.However, in the MgB_2 sample, the sintered temperature of 780°C with a holding time of 1 hour did not show superconductivity properties. Meanwhile, sintering temperature variation of 780°C, 800°C, 820°C with a holding time of 3 hours showed superconductivity properties, namely the appearance of Tc^{onset} and Tc^{zero} in each sample. The morphological structure and elemental composition were seen through SEM-EDS, where the surface morphology had a hexagonal grain shape. As the sintering temperature and holding time increase, the surface morphology of the grains becomes denser and more regular so that the porosity is smaller.

Keywords: magnesium diboride (MgB_2), stainless steel SS 316L, In-Situ powder in tube (PIT), sintering temperature and holding time, hot rolling, superconductivity.

1. INTRODUCTION

Nowadays, electrical energy has become one of the primary needs in daily life. Electrical energy becomes essential because it supports many aspects in daily life such as economic and health aspects. Electricity consumption has been increasing every year in Indonesia. According to The Ministry of Energy and Mineral Resource Indonesia (ESDM) in 2020, the annual increase in electricity consumption in Indonesia will continue until 2024. The electricity network in Indonesia commonly uses copper wire. Copper is one of the chemical elements which have conductor properties and corrosion resistant. Because of those reasons, wire or electric cable [1] uses 60% copper. The development of alternative material for copper is needed to balance the availability of copper in Indonesia.

Magnesium diboride (MgB_2) was founded in 2001 by Nagamatsu *et al* as superconductor materials. The critical temperature of $MgB_2$is higher than another metal oxide (Tc= 39 K) which can be functioned as cheaper material on the operational temperature in cryocooler (T = 20-25 K) [2]. MgB2 is a simple superconductor compound that consists of two metals unsure which have many unique properties such as high critical temperature (Tc ~39 K) above liquid helium, high current density of 106-107 A/cm2, and zero value of magnetic field in low temperature. The length of MgB_2 coherency is 3-12 nm which shows a higher value than high-temperature superconductor (HTSC) [3].

The development of MgB_2 material is expected to replace low temperature superconductors such as NbTi dan Nb3Snbecause of its higher critical temperature and abundant availability. In addition, it is also cheaper compared to NbTi and Nb_3Sn [4]. Erhan Aksu *et al.* revealed the effect of sintering temperature variation to the hexagonal microstructure of MgB_2 phase. The hexagonal microstructure of MgB_2 phase increases as the sintering temperature increases from 750°C to 950°C [5]. According to Akdogan et al, the sintering temperature of 850°C for 1 hour on MgB_2 superconductor wire showed high critical temperature (Tc^{onset} = 36,5 K) and the sintering temperature of 900°C for 1 hour showed higher critical temperature(Tc^{onset}= 37,1 K)[6].

Superconductor materials can be functioned as wires using powder in tube process with *in situ* methods. The advantage of *in situ* method is that the sample can be heated in atmospheric condition and that it doesn't require argon gas so it is simpler, cheaper, and higher in

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productivity level compared to other method [7]. In this experiment, stainless steel tube (SS 316) which has better chemical stability, better mechanical strength on high temperature and cheaper than Ni and Cu tube was used. The stainless-steel tube did not react with MgB_2 powder so that a better result of MgB_2 phase was obtained. The higher sintering temperature of 825°C with two hours of holding time showed good superconductivity properties (Tc^{Onset}=38.2) [8]

In this experiment, a variety of sintering temperature and holding time using hot rolling process is conducted to obtain superconductor wire. That variety was implemented to get the optimum parameter in SS/MgB_2 wire superconductor synthesis. The novelty of SS 316 as powder sheath material which has good formability characteristics and hot rolling fabrication is implemented to maintain the size reduction results without stress concentration and minimal defects.

2. EXPERIMENT DETAILS

The materials used in this experiment were Kanto Chemical Japan magnesium powder with 100 mesh size and 98% of impurity, amorphous Luoyang boron powder with 95% of impurity from Henan Province, China, and SS 316L stainless steel tube. The experiment started by preparing SS 316L stainless steel tube and 12 cm and 2 cm cover rod. The cover rod was used as the cover of the stainless steel tube. After that, the Mg and B powder were weighed to obtain stoichiometric composition with the ratio of 1:2. Each of powder was meshed using mortar agate for two hours until it became nanometer particle size. The weighed powder was then put in the SS 316L stainless steel tube and compacted using small wire until the stainless tube was full and solid. This process aims to obtain the denser and solid particle, minimize the reaction of air and powder when the sintering process takes place, and prevent the sample to evaporate and stuck in the tube. The sample was sintered using a muffle furnace by placing the sample on the cork and then the sample was heated on the muffle furnace. The sintering process aims to compress and diffuse the atom to form strong bonds between particles[9]. The sintering process was done in a variety of temperature of 780°C, 800°C and 820°C with holding time of 1 hour and 3 hours. All the samples started off at room temperature (T = 27 K) then they were sintered at 780°C for 2 hours and 36 minutes on 5°C/minute of heating rate (same value of heating rate on all samples). After that, the sample was constantly heated at 780°C for 1 hour for sample 1A. The same step was done to the other sample and the variation of sintering temperature and holding time can be seen in Table-1.

Table-1. Variation of sintering temperature and holding time in MgB_2 superconductor wire.

Sample	2^{nd} sinter (°C)	Hold time (minutes)
1A	780	60
1B	780	180
2A	800	60
2B	800	180
3A	820	60
3B	820	180

The samples were hot-rolled using wire rolling at T= 300°C to deform the samples into wire or cable [10]. The reduction of 50% from 8mm to 4 mm of tube diameter in all samples was applied in the hot rolling process. The reduction process was conducted to form the MgB_2 wire and prevent the samples from failure and cracking in the SS 316L stainless steel tube. The hot rolling process reduced the thickness and extended the wire to become longer than before (deformation process). The hot rolled and sintered samples were cut in 2 cm long in cross-section area using the conventional machine to simplify the characterization process. The cut samples were characterized using X-ray diffractometer (core sample), cryogenics testing, and SEM-EDS testing (cross-section).

3. RESULT AND DISCUSSIONS

3.1 MgB_2 Structural Analysis

The XRD characterization was applied to superconductor wire samples with a variety of sintering temperature (780°C, 800°C, 820°C) and holding time (1 and 3 hours), processed by hot rolling at 300°C with 50% reduction from 8 mm to 4 mm. The results of XRD characterization were analysed using Highscore Plus software using COD_Oct2014 database.

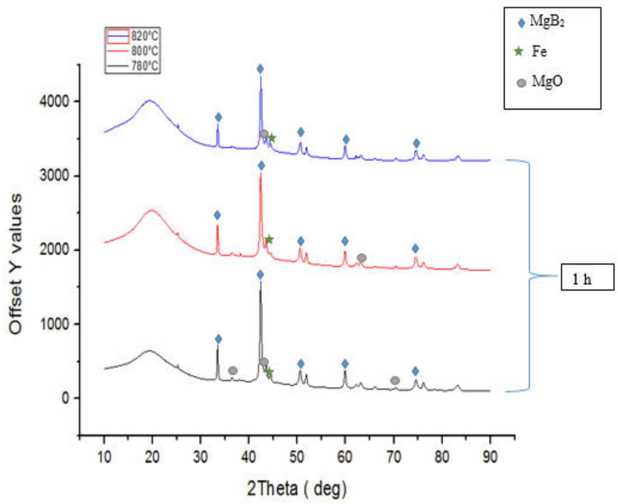

Figure-1. XRD pattern of MgB_2 wire with sintering variations and 1 hour of holding time.

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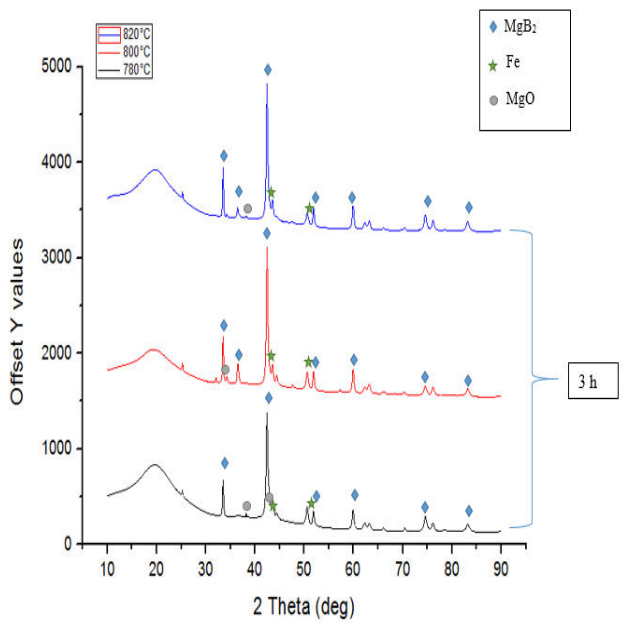

Figure-2. XRD pattern of combined MgB_2 superconductor wire using variation of sintering temperature and 3 hours of holding time.

According to Figure 1 and Figure 2, it can be concluded that MgB_2 is a dominant phase with database reference of ICDD 96-100-0027 which proved that Mg and B powder successfully synthesize and react well. In each samples, MgO phase was found which revealed impurities using database reference 96-900-6793, as well as Fe phase using database reference96-901-5072. However, the sample with 780°C of sintering temperature with 1 hour of holding time had lower content of MgB_2 phase and higher impurity phase than the other samples in Figure-1. Furthermore, the sample with 800°C of sintering temperature and 3 hours of holding time had higher content of MgB_2 phase and lower impurity content than the other samples in Figure-2. The formation of impurity Fe phase in every heating temperature came from the tube which were used as sample containers. Meanwhile, the impurity of MgO phase came from the oxidation reaction of Mg.

There is a chance for oxidation reaction because of the trapped air inside the SS316L stainless tube during the sintering process. It can be revealed form the Figure-1 and Fugure-2 that the increasing of sintering temperature could increase the dominant MgB_2 phase and reduce the impurity phase in the MgB_2 supercondctor wire. The XRD results showed the effect of sintering temperature to crystallinity of the sample which can be seen from the calculation of crystallite size using High Score Plus software.

Table-2. Crystallite size of MgB_2 sample with 1 hour of holding time using Scherrer Calculator in High Score Plus software.

Temperature	FWHM. [°2Th]	B std. [°2Th]	Peak pos. [°2Th]	B struct. [°2Th]	*Crystallite size* [Å]
780°C	0.310	0.008	42.377	0.302	282
	0.197	0.008	33.494	0.188	441
	0.472	0.008	59.872	0.464	198
		Average			307
800°C	0.300	0.008	42.370	0.292	292
	0.236	0.008	33.454	0.228	364
	0.394	0.008	59.896	0.386	238
		Average			298
820°C	0.300	0.008	42.400	0.292	292
	0.236	0.008	33.516	0.228	364
	0.394	0.008	59.910	0.386	238
		Average			298

Table-3. *Crystallite size* of MgB_2sample with 3 hour of holding time using Scherrer Calculator in High Score Plus software.

Temperature	FWHM. [°2Th]	B std. [°2Th]	Peak pos. [°2Th]	B struct. [°2Th]	Crystallite size [Å]
780°C	0.300	0.008	42.390	0.292	292
	0.197	0.008	33.514	0.189	439
	0.394	0.008	59.897	0.386	238
		Average			323
800°C	0.290	0.008	42.395	0.282	302
	0.197	0.008	33.497	0.189	439
	0.354	0.008	59.813	0.346	265
		Average			335
820°C	0.270	0.008	42.302	0.262	325
	0.236	0.008	33.498	0.228	364
	0.354	0.008	59.891	0.346	265
		Average			318

The sample with 800°C of sintering temperature with 3 hour of holding time hadbigger size of crystallite than sample with the same sintering temperature with 1 hour of holding time. This phenomenon was caused by the lower value of FWHM as the bigger size of crystallite [11].

3.2 Electric Properties of MgB_2

The resistivity measurement is conducted to find the electric properties of MgB_2superconductor shown by the critical temperature (Tc).

VOL. 17, NO. 7, APRIL 2022 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

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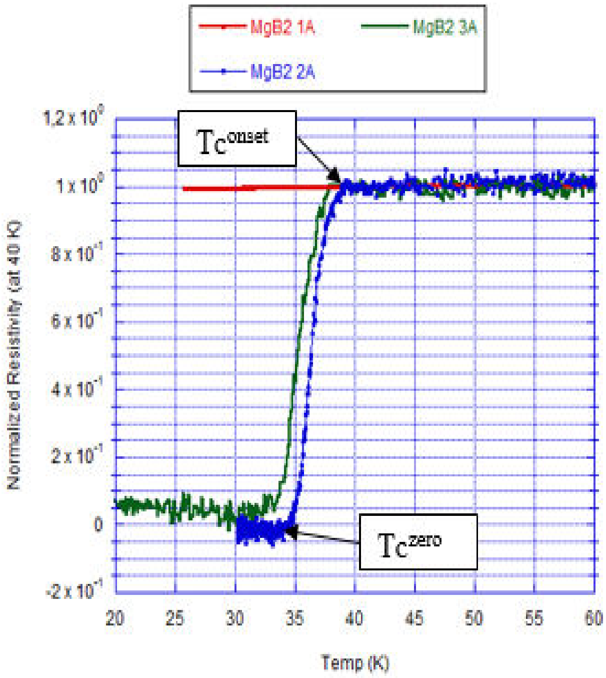

Figure-3. Resistivity vs temperature of MgB_2 sample with variation of sintering temperature and 1 hour of holding time.

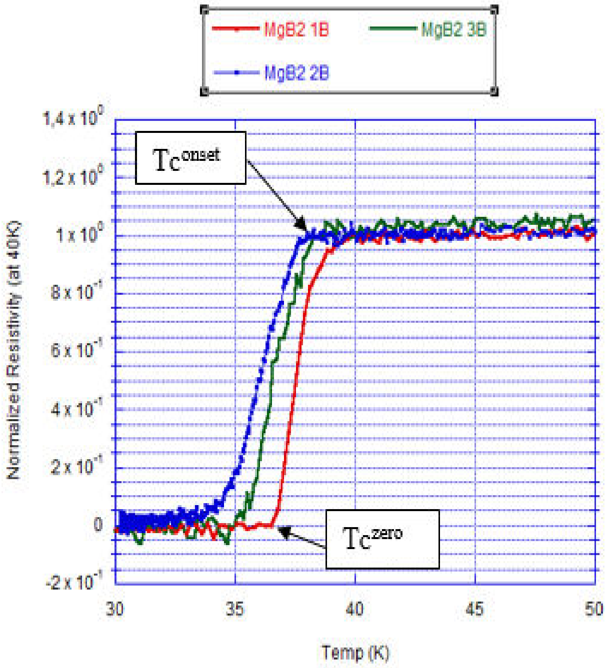

Figure-4. Resistivity vs temperature of MgB_2 sample with variation of sintering temperature and 3 hours of holding time.

Figure-3 showed that the sample with sintering temperature of 780°C didn't have superconductivity curve and tended to be non superconductor material. This phenomenon was caused by the high content of impurity phase such as MgO and Fe, the high percentage of oxide atom, and the lack of holding time in sintering process. The other factor was the porosity in the MgB_2 samples that caused the unstability of electric current when measuring resistivity. The sample with 800°C of sintering temperature showed Tc^{Onset} of 39.55 K and Tc^{Zero} of 34.69 K. Furthermore, the sample with 820°C of sintering temperature showed Tc^{Onset} of 38.44 K and Tc^{Zero} of31.30 K which showed a lower value from the previous sample. This phenomenon was caused by the impurities contained in the samples. Qian Zhao et al indicated in their research that the decreasing of Tc value is caused by the impurity phase [12].

Table-4. The value of Tc^{Onset} and Tc^{Zero}on MgB2 superconductor wire with variation of sintering time and 1 hour of holding time.

Temperature	Tc^{Onset} (K)	Tc^{Zero} (K)	ΔTc (K)
780°C	-	-	-
800°C	39.55	34.69	4.86
820°C	38.44	31.30	7.14

Table-5. The value of critical transition sample.

Temperature	Tc^{Onset} (K)	Tc^{Zero} (K)	ΔTc (K)
780°C	39,20 K	36,50 K	2,76
800°C	38,55 K	32,50 K	6,05
820°C	39, 16 K	34,04 K	5,12

It can be concluded that the variation of sintering temperature and holding time could affect superconductivity properties such as critical temperature (Tc).

3.3 The MgB_2 Morphological Analysis

The SEM-EDS characterization revealed the morphology of MgB_2 wire surface with variation of sintering temperature and holding time using 10.000x magnification (left) and mapping (right) which can be seen on Figure-5 and Figure-6.

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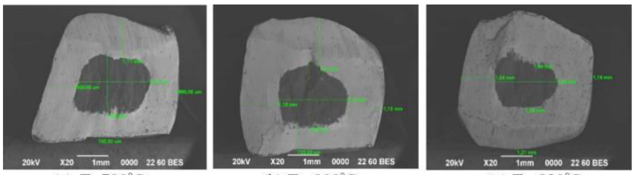


(a) T=780°C (b) T= 800°C (c) T= 820°C

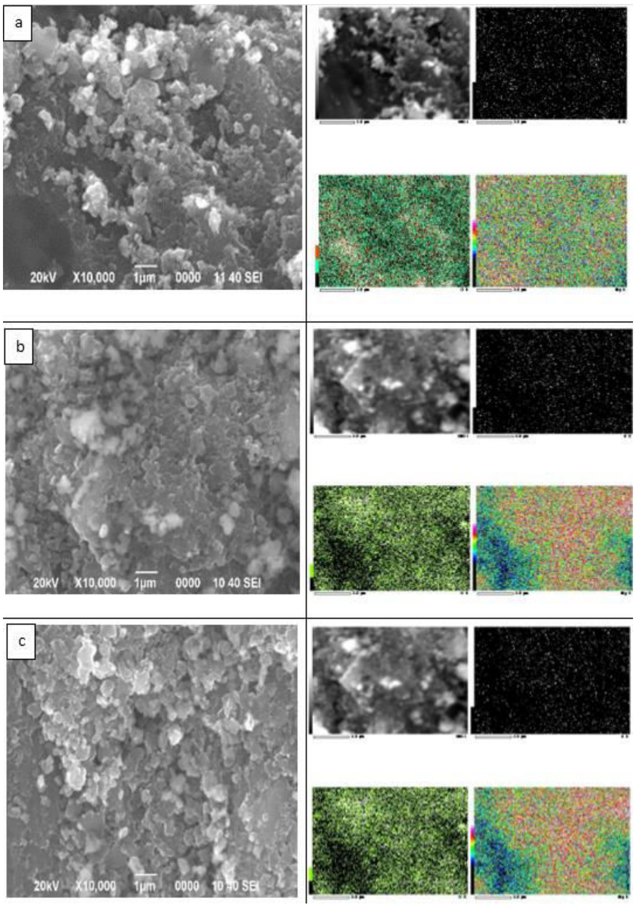
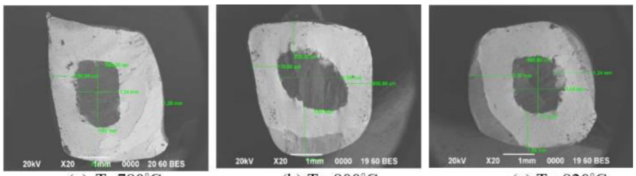


Figure-5. Morphology of MgB_2 wire surface with variation of sintering temperature and 1 hour of holding time.



(a) T=780°C (b) T= 800°C (c) T= 820°C

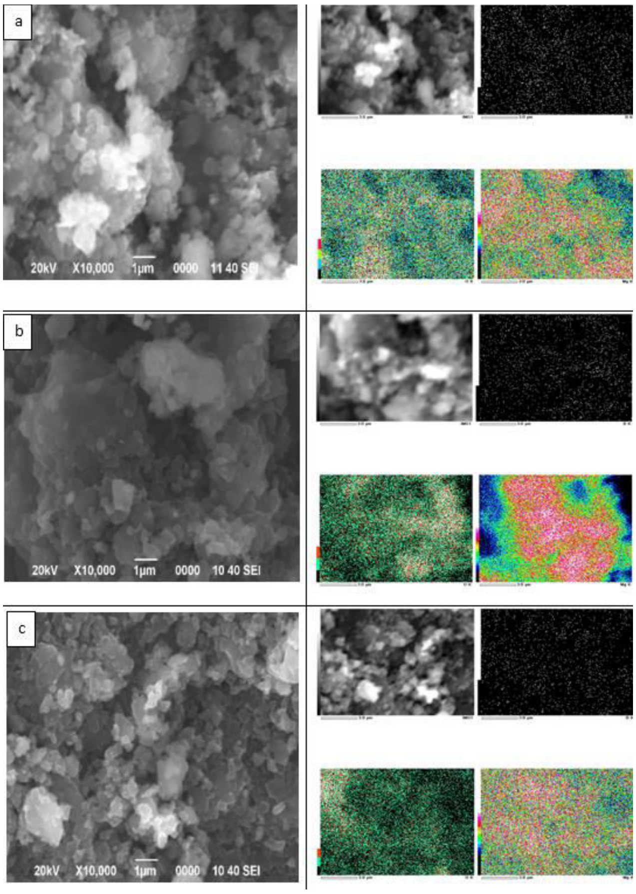


Figure-6. Morphology of MgB_2 wire surface with variation of sintering temperature and 3 hours of holding time.

According to Figure-5 and Figure-6 as the results of SEM-EDS characterization, it can be seen that the morphology of MgB_2 wire surface shape began to form a homogeneous solid shape such as small hexagonal grain. The hexagonal grain shape is the characteristic of MgB_2 particles which are synthesized using powder metallurgy [12]. An increase in sintering temperature and holding time affected the morphology of MgB2 grain. An increase in sintering temperature and holding time could increase the agglomeration of MgB_2 grain and thickness of the hexagonal grain shape. The grain bound each other and created agglomeration. The agglomeration of MgB_2 grain would get denser as the sintering temperature and holding time increase, which can be seen in Figure-5. The increasing of holding time affected the size of MgB_2 grain, in which a higher holding time created a bigger grain size, as seen in Figure-6.

The EDS mapping with 10.000x magnification in the right side of Figure-5 and Figure-6 showed that all samples have different grain densities. A higher sintering temperature and holding time would make the grain denser, thicker, and more homogenous than the other sample. The EDS mapping also indicated the O element in all MgB_2 superconductor wire samples which means that the sample was oxidized with oxygen elements.

4. CONCLUSIONS

The synthesis and fabrication process of MgB_2 superconductor wire with a variation of sintering temperature (780^{o}C, 800 ^{o}C, 820 ^{o}C) and holding time of 1 and 3 hours were done successfully. The method used was hot rolling to produce wire, and the powder metallurgy used was in-situ powder in tube (PIT) that loaded Mg and B powder in the SS316L stainless tube. Cryogenic results showed that the resistivity value in the transition curve decreased drastically to 0 ohm and had Tc^{onset} and Tc^{zero}. The highest value of Tc^{onset} is the sample sintered at 800^{o}C and 1 hour holding time of 38.44 K. It can be concluded that hot rolling with higher value of sintering temperature and holding time will show better Tc value and superconductivity properties compared to other samples. The XRD characterization results showed the increase of MgB_2 phase and decreasing of MgO phase occurred as the sintering temperature and holding time increased. The crystallite size in 1 hour of holding time sample was smaller than the crystallite size in 3 hours of holding time sample. All the MgB_2 superconductor wire samples were likely crystalline. According to the SEM-EDS results, the microstructure of the surface morphology had the hexagonal grain shape for MgB_2 superconductor wire samples. An increase in sintering temperature and holding time caused denser and more homogeneous grain, resulting in lower porosity than another sample.

ACKNOWLEDGEMENT

The authors are grateful for the PUTI Doctor UI 2020-2021, under contract No. NKB-680/UN2.RST/HKP.05.00/2020 for their financial support. Gratitude is also addressed to the researchers and technicians of the superconducting team, as well as a research collaboration with the Universitas Sumatra Utara.

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