

The Effect of Holding Time Variations in MgB_2 Superconductor with Addition Nickel Prepared by Powder in Sealed Tube Method

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Abstract. In this research, the effect holding time on the synthesis of MgB_2 superconducting material with the addition of nickel has been carried out using the powder in sealed tube method. MgB_2 is a superconductor which has a critical temperature of ~ 39 K. The addition of 0%, 4%, and 8% Ni was added to study its effect on the superconductivity characteristics of MgB_2 . The sample preparation process begins by weighing the raw materials in the form of Mg, B and Ni powders according to stoichiometric calculations. The material is then crushed for 1 hour using mortar agate then put into a stainless steel tube SS 316L and the PIST pressing process is carried out with a pressure of 5 MPa, then the sintering process is carried out using a muffle furnace with a temperature of 800°C and holding time for sintering temperature for 1 hour, 3 hours and 5 hours. Samples were characterized by means of XRD, SEM-EDS and Cryogenic Magnet. The results of phase identification through XRD showed that the phases formed were MgB_2 , $\text{MgNi}_{2.5}\text{B}_2$, MgO , Mg and Ni. Morphological structure and elemental composition were seen through SEM-EDS. From the Cryogenic Magnet test, pure MgB_2 samples with a holding time of 1 hour had a critical temperature T_c of 36.29 K and did not have T_c . Meanwhile, pure MgB_2 at a holding time of 5 hours has a T_c of 42.18 K and a T_c of 25.55K.

Keyword: superconducting, magnesium diboride, nickel doping, porosity.

Received 18 November 2019 | Revised [10 December 2019] | Accepted [14 February 2020]

1 Introduction

Based on data reports on world nickel production and reserves from USGS (2015), Indonesia is one of the countries capable of producing nickel in the world with lateritic types. In terms of potential reserves, Indonesia is in the sixth position with nickel of 5% from total nickel reserves in the world. Nickel, which is one of the main class elements of stainless steel, has experienced development with the increasing demand for stainless steel [1]. As stated by Moskalyk (2002)

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that more than 65% of nickel is currently used in the stainless steel industry and 12% is used in the super alloy manufacturing industry. [2]. Magnesium diboride (MgB_2) is superconductor material with temperature T_c increase $\sim 39\text{K}$ [3-4]. The MgB_2 material began to develop in the 1950s, but Nagamatsu, et al discovered its superconductivity in 2001. Nagamatsu et al, revealed that MgB_2 is a complex compound and consists of two metal elements that have superconducting properties, so scientists are interested in expanding the superconducting material. Besides having a critical temperature above liquid helium, MgB_2 has a high critical current density of $106\text{-}107\text{ A/cm}^2$ and has no magnetic field when the temperature is low. The development of the MgB_2 material is expected to be able to replace the Nb_3Sn material, because MgB_2 has a higher critical temperature and a cheaper price compared to Nb_3Sn material [5-6].

In a study conducted by O.F de Lima et al (2010) which states that pure phase MgB_2 with the addition of Ni can increase the critical current density values. With variations in the Ni composition of 0.5%, 1%, 2%, 3%, and 5% with a temperature of 800°C sintered for 5 hours, giving the best results by increasing the values of critical current density at the 0.5% addition of Ni composition [7]. According to research conducted by Zhao et al (2009), a sample of MgB_2 which is added too much Ni will have a higher resistivity so that it tends to reduce superconducting properties compared to MgB_2 sample [8].

In this article, the effect holding time of sintering temperature and the weight percent of nickel doping in the manufacture of superconducting materials so that the optimal holding time parameters are expected in the synthesis of MgB_2 superconducting material using the powder in sealed tube method and evaluating the effect of adding nickel doping on crystallinity and microstructure of MgB_2 .

2 Materials and Methods

The materials used were Magnesium powder with a size of 100 mesh, (purity: 98% Aldrich), Boron powder with a size of $1\text{ }\mu\text{m}$, (purity: 92.1%, density: 2.34 g/mL , brand KGaA Darmstadt Germany) and Nickel powder (Ni) $10\text{ }\mu\text{m}$, and stainless steel tube type 316L. The process begins with the preparation of SS 316L tubes by cutting each tube with a length of 6 cm and then continuing the cleaning process using ethanol to minimize tube dirtiness before the material enters it. After cleaning, the tip of the tube will be compacted before inserting the sample. Furthermore, the powder is weighed with a composition that has been determined according to stoichiometric calculations. The variations in the addition used were Ni powder with 3 variations in the weight composition of each addition of 0; 4%, and 8% by weight. The powder mixture is then crushed using a mortar agate for 1 hour until it becomes homogeneous. Then proceeded with the process of entering the powder into the SS 316L tube and then the sample is compacted with a pressure of 5 MPa using a hydraulic compacting machine (Hydraulic Press). This process aims to make the sample particles tightly and densely arranged

so there is no reaction between air and the sample and when sintered the sample does not evaporate and is stuck in the tube [9].

The sample is then proceeded to the sintering process using a muffle furnace by placing the sample on the cork as a sample container, then the sample is put into a heating furnace until it lies in the heating area. This process aims to make the sample more compressed and it is hoped that the atomic diffusion process will occur and form strong bonds between particles [10]. The final sample result is in the form of a chunk or core sample. The sintering process was carried out by varying the holding time for 1 hour, 3 hours and 5 hours for each sample at a rate of 50°C/minutes from the initial temperature of 27°C (room temperature) for 2 hours 50 minutes then kept constant at 800°C and then the sample was left cold in the furnace. The results of the sintering are then carried out characterization using X Ray Diffractometer (XRD), Scanning Electron Microscopy/Energy Dispersive Spectroscopy SEM/EDS and Cryogenic magnet to determine the phases that appear through the diffraction pattern, the microstructure morphology formed and the superconductivity properties characterized by measuring resistivity of the material.

3 Result and Discussion

3.1 Structures Analysis of MgB_2

The effect of nickel addition on the formed phase is shown in Figure 1. The results of the XRD analysis with the diffraction pattern that emerged from the three variations of % Ni addition in the three sinter temperature holding times for 1 hour, 3 hours, and 5 hours.

From the three variations in addition of Ni at each holding time of the sinter temperature from the diffraction pattern in Figure 1, it is found that the MgB_2 phase is the primary phase or the main phase with the ICSD 96-100-0027 reference database formed which proves that Mg and B powders were successfully synthesized and reacts well. Furthermore, in the diffraction pattern with the addition of Ni with variations in the addition of Ni 0%, 4% and 8% wt, the secondary phase of $\text{MgNi}_{2.5}\text{B}_2$ was obtained with a reference database 96-720-9438 where this phase is a reaction between Ni with Mg and B and can be obtained above 600°C. Furthermore, the MgO phase and the Mg element, which are the impurities, are also obtained. The MgO phase with reference database ICSD 96-900-6458 reacts with Mg to become MgO and it can also occur due to the compacting process on SS 316L which is less dense so that there is still a gap for air trapped in it. The Mg element itself is due to the presence of Mg which has not yet completely melted during the sintering process in the SS 316L tube. Of the three variations, it can be seen that the greater the % addition of nickel is added, the more secondary phase $\text{MgNi}_{2.5}\text{B}_2$ is formed and there is no variation of 0% Ni or without addition. This indicates that this phase arises from the addition of nickel powder.

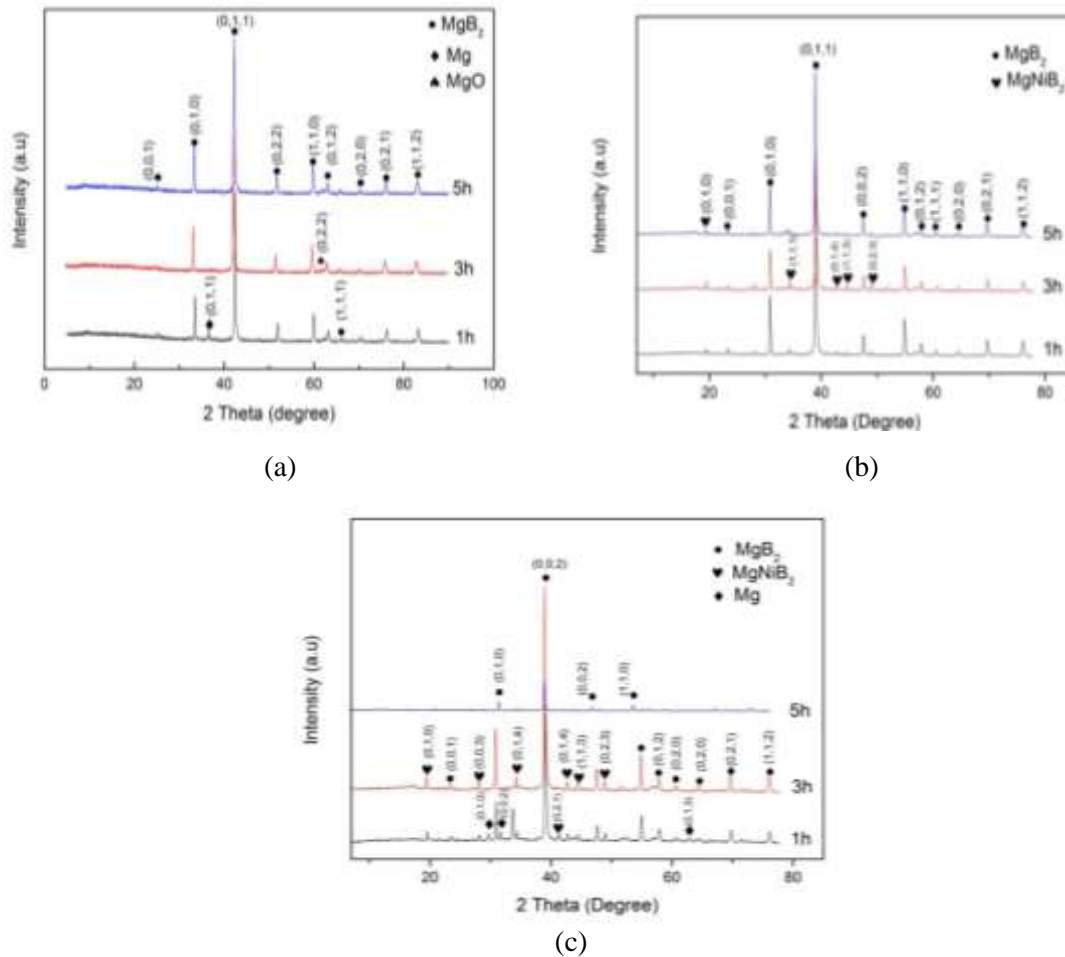


Figure 1. XRD Patterns with addition of Ni (a) 0%, (b) 4%, (c) 8 wt% with holding time for 3 hours

From the XRD data analysis, it was also seen the effect of the addition of Ni on crystallinity which can be seen by calculating the crystallite size using High Score Plus (HSP) software.

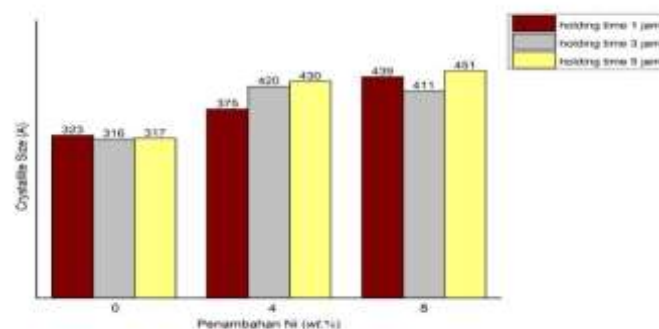


Figure 2. Histogram of crystal size (Scherrer Calculator) on the variation of holding time

The results of the calculation show that in samples with a sintering temperature of 800°C and a holding time of 5 hours of sintering temperature with the addition of Ni tended to increase the crystallite size, but in samples without the addition of Ni the largest crystal size was obtained at

1 hour holding time. A significant increase in crystal size with increasing holding time of sinter temperature occurred in the sample with the addition of Ni by 4%. This is because the FWHM is getting smaller so that the crystal size is getting bigger [11].

3.2 Morphology Analysis of MgB_2

Characterization using SEM-EDS can be seen the effect of Ni addition and the variation of holding time in sinter temperature on the surface morphology and microstructure of the superconducting MgB_2 as shown in Figure 3 and Figure 4.

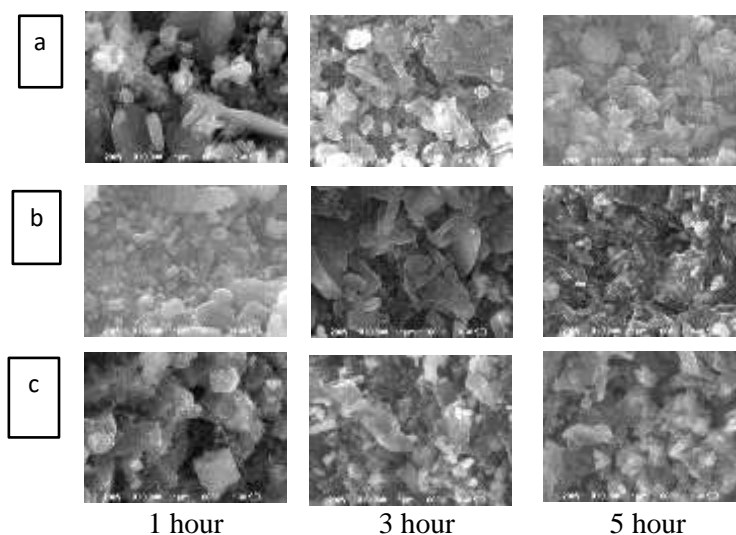


Figure 3. SEM of MgB_2 with addition Ni variations (a) 0%, (b) 4%, and (c) 8%

Figure 3 shows the SEM observations with magnification of 10000X with a temperature of 800°C and holding time of the sinter temperature, where the surface morphology shows its characteristic, which is in the form of clumps that bind with each other as the holding time of the sinter temperature increases. In the picture it is also seen that the sample with the addition of Ni shows white spots which indicate the presence of Ni powder that has combined with Mg and B powders [12].

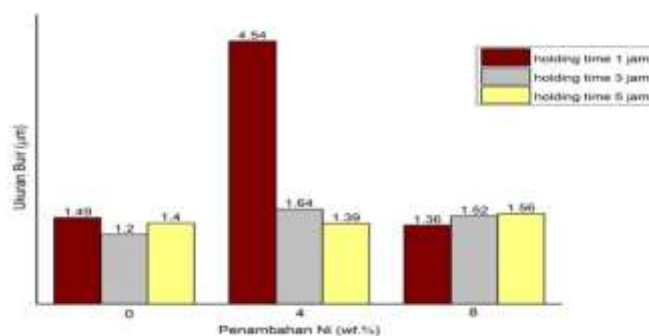


Figure 4. Histogram of particle size distribution on holding time variations and Ni addition

Figure 4 is the calculation data for the average grain size of the MgB_2 superconductor. From these data, it was found that the samples that experienced a significant increase in grain size

along with the increase in the holding time of the sinter temperature occurred at the addition of 8% Ni. However, the largest grain size occurred in samples with the addition of 4% Ni and a holding time of 1 hour, the rest decreased with the increase in the holding time of the sinter temperature.

3.3 Electric Properties of MgB_2

The resistivity measurement to know electric properties in MgB_2 superconductivity which is indicated by the critical temperature (T_c) shown in Figure 5.

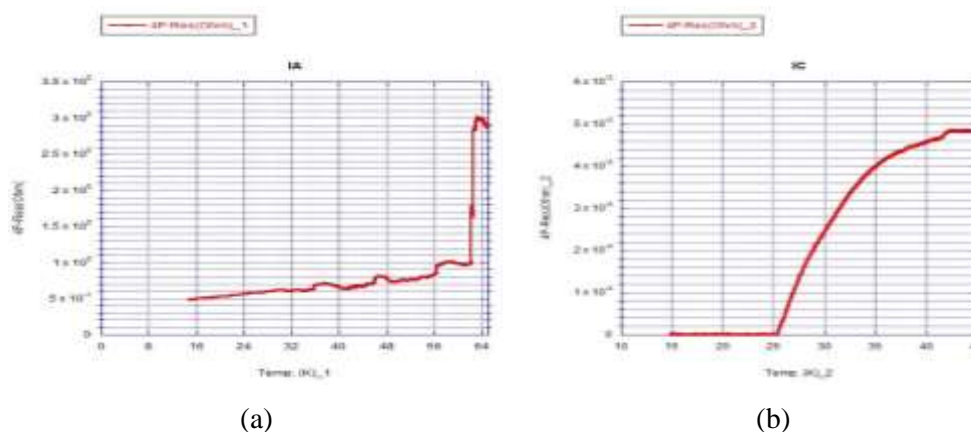


Figure 5. Resistivity on MgB_2 holding time variations with MgB_2 and without Ni additions (a) 1 hour, and (b) 3 hour

Figure 5 can be seen that at temperature of 800°C without the addition of Ni with a holding time of 1 hour shows the critical temperature value of T_c Onset obtained is 36.29 K but T_c Zero is not achieved due to a very hollow sample and lots of impurities. Whereas at the 5 hour holding time, the T_c Onset obtained was 42.18 K and T_c Zero was 25.55 K.

4 Conclusion

Synthesis of MgB_2 material with the addition of Ni using the powder in sealed tube method was successfully carried out. Based on the results of the cryogenic magnet test, the two samples have superconductivity which is indicated by a drastically dropping transition curve. Based on the XRD test results of the superconducting MgB_2 with the addition of Ni and the variation of holding time, it was found that the primary phase was MgB_2 and the secondary phase $\text{MgNi}_{2.5}\text{B}_2$, as well as compounds in the form of MgO and Mg elements as impurities. The best crystal size increase in the sample with the addition of 4% Ni along with the increase in the holding time of the sinter temperature. Mg-B-Ni grains were evenly distributed in the superconductor as the holding time of the sinter temperature increased and the amount of Ni added. The best grain size occurred in the sample with the addition of 4% Ni and a holding time of 1 hour. Based on the cryogenic magnet test, the sample without the addition of Ni with a holding time of 1 hour shows the critical temperature value of T_c Onset obtained is 36.29 K but

Tc Zero is not achieved, while at the holding time of 5 hours the Tc Onset obtained is 42.18 K and Tc Zero of 25.55 K.

Acknowledgement

This research supported by Universitas Sumatera Utara (USU) Medan, Indonesia and Research Centre for Metallurgy Indonesian Institute of Sciences (LIPI), Tangerang Selatan, Banten, Indonesia.

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