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Development of magnetic materials for cobalt titanium oxide using mechanical milling and the characterization methods

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Abstract

In the synthesis of magnetic materials for absorbing radar waves, it is necessary to have a base material that has unique magnetic properties that can reach the broadband characteristics of radar frequencies. Cobalt titanium oxide is synthesized using mechanical milling. The phase formed by calcination at 1000°C with a holding time of 5 hours. Sample characterization was carried out by XRD, VSM and VNA. Based on the research results, it was found that the material obtained had semiparamagnetic properties. From the SEM measurements, it is reactive to the sintering temperature. Radar wave absorption measured by VNA shows that cobalt titanium oxide shows a value with uniform absorption in the X-band wave.

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INTRODUCTION

Material technology is made or composed from several raw materials that are used to make products that are more useful, especially the development of electronics and wireless information technology. One is that the development of radar absorbing materials with advanced microwave properties has emerged as a non-negligible concern (Jia, et al., 2018). There is a growing level of interest in hexaferrites, and it is still growing today. They have been produced on a large scale and are an important, commercial and technological material used in many electrical systems, such as permanent magnets, magnetic recorders and data storage devices (Pullar, 2012; Koutzarova 2009). One that is being developed is a radar wave absorbing material that can be developed on aircraft, coatings, or military uniforms. In this study, the preparation of materials by making raw materials that have conductivity and electricity that can be absorbed in electromagnetic waves. Achieve these properties, the manufacture of the material has a magnetic material belonging to the ferrite material group (Jiang et al., 2008).

Many magnetic studies are devoted to many things, especially on the interaction between materials and magnetic fields, the potential energy that is owned, by the magnetic material that is

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owned. The interaction observed through changes in the magnetic moment or magnetic force experienced by the sample due to the distribution of the magnetic field. Therefore, the materials chosen are materials that have very high permittivity and permeability properties (Nasution & Astuti, 2012), namely materials that have magnetic and electrical properties as the basis for wave absorption. These materials are the result of material engineering which will have two properties, namely having permittivity and permeability properties to equalize the wave phase (resonance) with radar waves so that they can interact with each other resulting in the absorption of radar waves. Preparation of Cobalt Oxide doped with titanium to produce increased physical properties (magnetic properties).

Cobalt metal is a metal that has an important role and is an essential metal element. Cobalt metal is usually used mixed as an alloy metal. Besides that, titanium oxide is an oxide compound that has a white color, is a good catalyst material, as well as corrosion resistance and good electrical conductivity in reaction conditions.

Nano Titanium Dioxide (TiO2) has been extensively studied in recent years due to its high applications as photovoltaic batteries, semiconductor photocatalysts, and gas and humidity sensors. Cobalt titanium oxide can be used as a paint material that is used to coat the surface of a material with decorative (beautifying), reinforcing or protective purposes (Weldon, 2009). This section includes metal coating (electro coating, galvanizing), pastic coating, paper coating, powder coating and the paint itself. So, this paint is part of a bigger one, namely surface coating.

This study was conducted to determine the material characteristics of the Co3O4 material, and TiO2 then formed a new material to make impedance matching waves occur which functioned by engineering from conductive and capacitative materials.

METHODS

Materials : The materials used in this experiment are Cobalt Oxide, Titanium Oxide, Ethylene.
Tools : The tools used in this experiment are ball mill, measuring cup, mould, furnace, digital balance, and mechanical milling.
Preparation : Weighing the cobalt formula added with ethylene is then milled using the solid reaction method using mechanical milling for 6 hours. Then the material was sintered using a Thermolyne 6000 furnace at 1000℃ for 5 hours. Furthermore, the material is characterized using X-ray diffraction (XRD), Scanning Electron Microscope (SEM), Vibrating Sample Magnetometer (VSM) and Vector Network Analyzer (VNA) for testing radar wave absorption. (The research process can be seen in Figure 1)

Cobalt Oxide and Titanium Oxide materials were synthesized using the solid reaction method through mechanical milling techniques. The mixed material is determined based on stoichiometric calculations then weighed and put into a stainless vial. Then the mixture was added with a ball mill and ethanol in a ratio of 1:2. The process was milled for 5 hours using High Energy Milling, then dried at 100° C in an oven. The dried samples were pulverized and sintered at 1000° C in a furnace. The X-ray diffraction pattern of the modified sample was measured with an X-Ray Diffractometer (XRD) using CuKα radiation (λ = 1.5406 Å). The phase formed from the sample was analyzed using the Match Program and General Structure Analysis (GSAS) qualitatively and quantitatively. After that, the surface morphology and sample components were observed by Scanning Electron Microscope (SEM) and Vibrator Network Analyzes and Vector Network Analyzes.

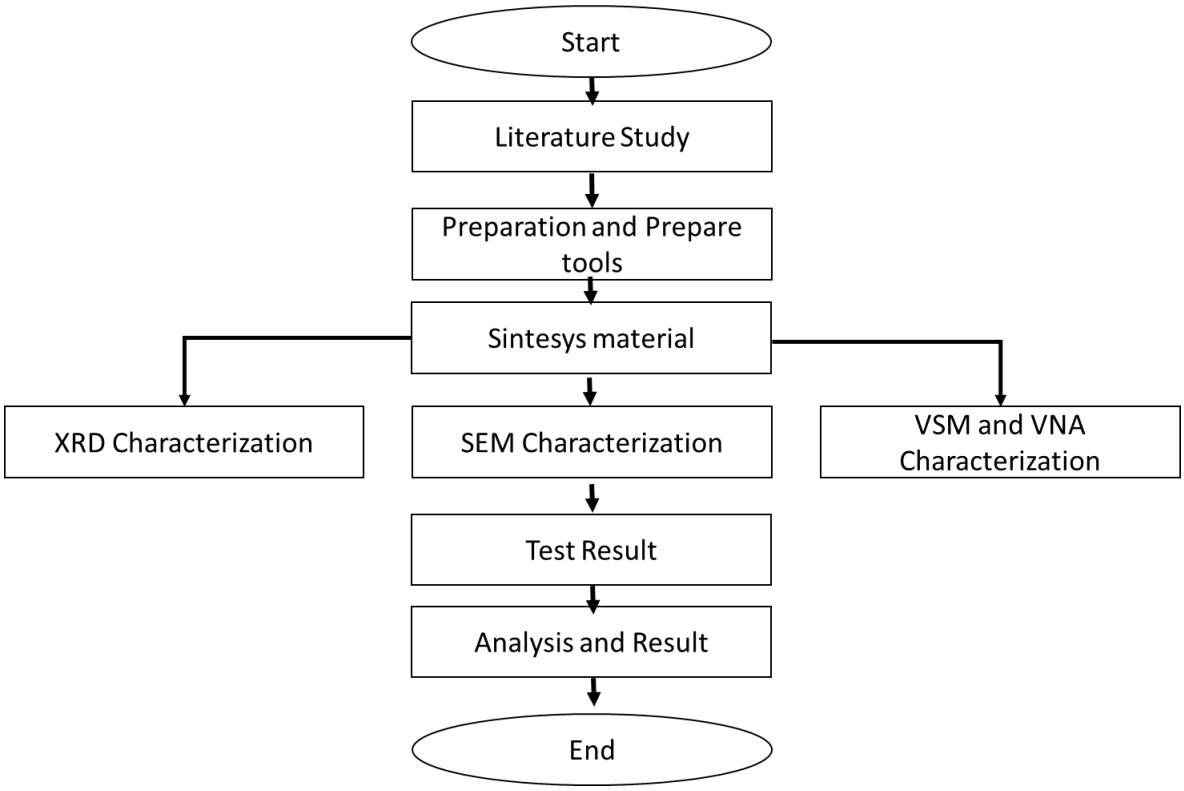


Figure 1. Flow of absorber material synthesis

RESULTS AND DISCUSSION

This experiment aims to determine the ability of raw materials to have absorber properties on radar waves. Figure *2* shows the result of the process of making wave absorber materials with the main ingredients being cobalt oxide and titanium oxide which have magnetic and electrical elements to increase the permeability and permittivity coefficients for radar wave absorbing materials. In terms of color, the resulting powder forms a green pigment, besides being used as a military camouflage color for its field of operation, namely tropical forests, it can also be used as a paint pigment on buildings or vehicle bodies.

Figure 2. Manufacturing Result Material of $CoTiO_3$

Results of qualitative and quantitative analysis with X-Ray Diffraction Characterization

Qualitative analysis of XRD aims to identify crystal structures in the form of solids from the diffraction patterns formed where there is interference from the X-ray construction. From the data the $CoTiO_3$ material appears in the diffraction patterns representing crystal planes for matching COD

data with quantitative analysis using the GSAS program. The milling process creates a single phase which means the ingredients are mixed homogeneously. To find out the phase formed in the magnetic material that has been made, X-ray diffraction measurements were carried out using an X-Ray Diffractometer.

The qualitative analysis of XRD aims to identify the crystal structure in the form of a solid from the diffraction patterns formed where there is interference from the X-ray construction. From the data, the $CoTiO_3$ material that appears in the diffraction pattern represents the crystal plane for matching COD data. Qualitative and quantitative analysis referred to Open Database Crystallography with card numbers (JCPDS: 99-100-8694), and (JCPDS: 99-200-3340) for $CoTiO_3$ phases, respectively.

Analysis with reference to the material crystallography of cobalt oxide and titanium oxide formed a phase to $CoTiO_3$. A complete summary of the results of the refinement of the X-ray diffraction pattern from the $CoTiO_3$ material is shown in Table 1. The density is carried out to determine the density value of the sample that has been made, the table shows the value (ρ 4,987 gr.cm-3) which shows that in the presence of titanium substitution then the density value has a lot of substitution effect on hexagonal phrase bonds, it can be seen from Figure 3. $CoTiO_3$ *Crystal structure model* The material that has been made proves that titanium substitution does not change the phase or crystal structure which indicates that the substitution process can be carried out.

Table 1. Structural parameter values in materials

X	Fasa	Parameter Kisi (Å)			V (Å³)	ρ (gr.cm^{-3})	Fraksi (%)	Rwp	χ^2
		a	b	c					
0	$CoTiO_3$	5.0660(0)	5.0660(0)	13.9169(9)	309.31(9)	4.987	100	14.54	1.085

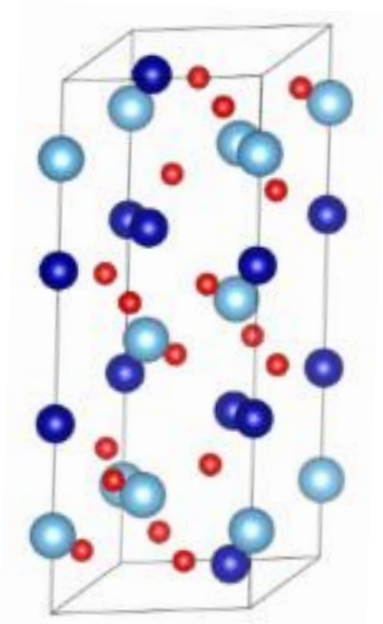
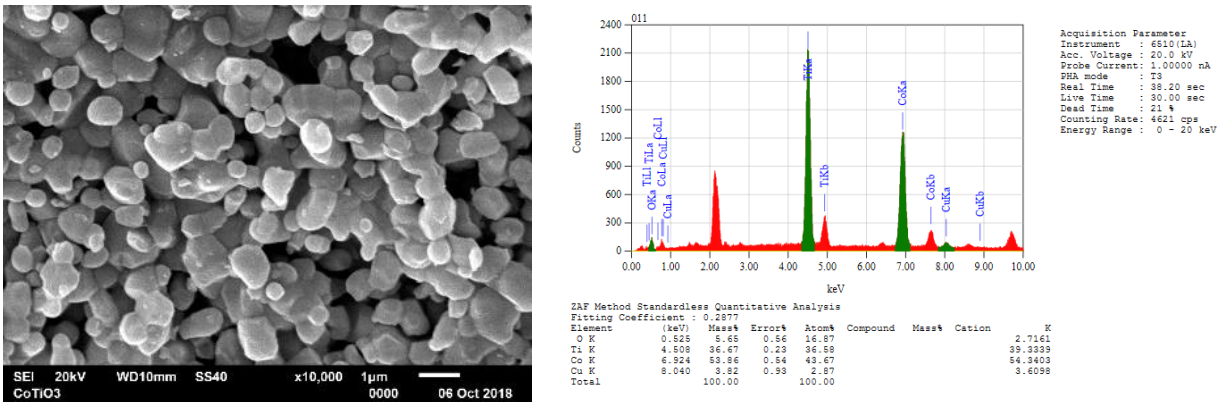


Figure 3. $CoTiO_3$ Crystal structure model

Results of Particle Morphology Analysis with SEM-EDS Characterization

Conventional characterization techniques to obtain information to provide "vision" assistance for researchers to be able to see the appearance around the interface between materials and the formation of images with objects that are relatively small by providing relatively low contrast at high magnification.



(a) (b)

Figure 4. SEM data

Figure 4(a) shows the morphological structure of the $CoTiO_3$ material. The results of the $CoTiO_3$ material have a homogeneous shape and particle size between one another. While other compositional variations can be seen in Figure 4(b), it appears that the particles have identical sizes and shapes. By looking at the SEM and EDX data, in terms of size and component the material has the same data, namely having pure material formed $CoTiO_3$ which is one phase and homogeneous, the synthesis that is made does not appear other impurities. The results of the refinement are also supported by the observation of the surface morphology of the particles by SEM shown in **Figure 4**. Uniformity is relatively even across all samples with particle sizes reaching micrometers. In this case, if it is related to the GSAS data, it succeeded in forming a single phase which is relatively the same size, and the two materials are substituted for one another.

Results of Magnetic Properties Analysis with VSM Characterization

VSM testing is carried out to determine and determine the magnetic properties of materials, which provide information data regarding magnetic properties due to changes in the external magnetic field which are described in the hysteresis curve. The curve that is owned by the $CoTiO_3$ material in the explanation of the curve can be expressed in terms of the magnitude of the remanent magnetization. The magnitude of the magnetic remanence describes the residual magnetism that a material has when external forces are removed or made zero. Based on Figure 5, the shape of the hysteresis curve for the magnetic domain belongs to the size of crystallite (a magnet that belongs to a very small size (nano) in the form of a group (cluster) which has paramagnetic properties, which causes the force interaction between atoms to be sufficient to change the direction of magnetization. This process causes the material to experience saturation magnetization. Although the hysteresis curve is small. Saturation magnetization becomes superparamagnetic when given a field strength of 60 kOe at 0.82 emu/g.

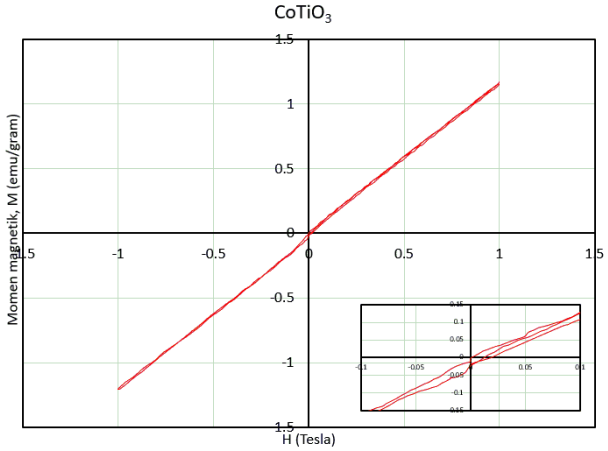


Figure 5. Analysis of VSM.

Table 2. Parameter magnetic $CoTiO_3$

	PARAMETER MAGNETIC		
SUBTANCE	Ms (emu/gr)	Mr (emu/gr)	Hc (Oe)
$COTIO_3$	0.1	0	0.82

A large magnetic remanence value is a must for a material, provided that the material represented has a radar absorbing material (Adi, 2010). In Table 2 it can be seen that the value shows that to form a good hysteresis value, on the other hand, having a high remanence magnetic material, the coercivity value is suggested to be smaller, close to zero, because the lower the coercivity value, the lower the remanence value that can be removed. Both of these reasons provide an explanation for the material having anomaly, possibly because the material does not have true ferromagnetic properties. This fact reflects the poor quality of the sample, because magnetic irreversibility is often present in magnetic ceramic samples

Results of Radar Wave Absorption Analysis with VNA Characterization

Testing the wave absorption value (reflection loss) is a comparison of the number of absorbed wave fractions due to wave resonance. The atoms that vibrate in the material have the same frequency as those that are vibrated through the VNA instrument. On the VNA device there are 2 outputs, namely S21 and S11, each of which sequentially means transmission loss and reflection loss so that in this study only took the S11 value.

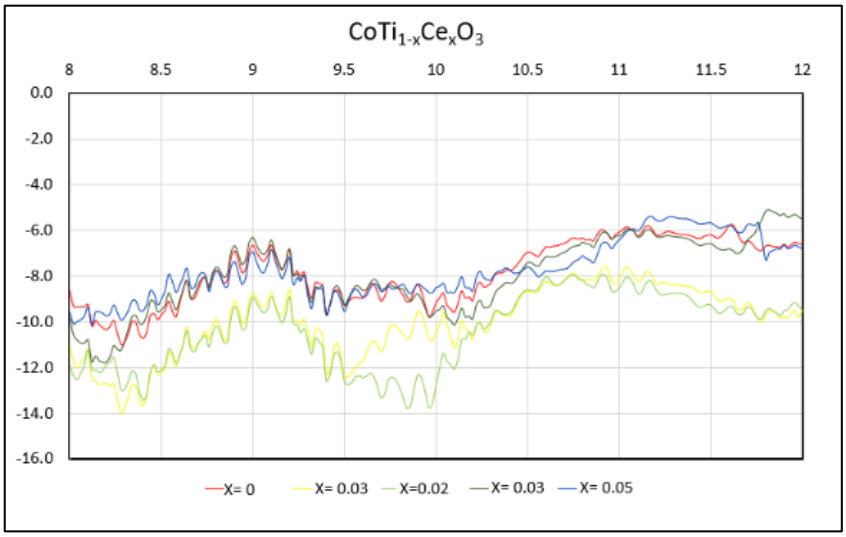


Figure 6. Value reflection loss

Because the concept requirement needed for radar absorbing materials (RAM) is the presence of the material's intrinsic properties (materials that have magnetic loss and dielectric loss). Figure 6 shows the magnitude of the frequency attenuation in reflection loss has the definition of the ratio of wave reflection in impedance values that occur in the frequency range 8-12 GHz at a material thickness of 2 mm. Based on the VNA results, the reflection loss is at a value of 6-10 dB with a frequency of 8-12GHz.

In theory, absorbent materials are divided into 2 factors, intrinsic and extrinsic factors. In the intrinsic factor, the material has a material that has low permittivity and permeability properties, so that microwaves are not easily transmitted properly by the material. Knowledge of permittivity and permeability values over a wide frequency range is an important input for this design calculation (Saptari, 2015). A brief survey of common classes of commercially available radar absorbing materials was made, with a number of references to specific materials (Gaylor, 1989). The decrease in magnetization properties causes the absorption ability of a material to also decrease. In this study, it has the same average reflection loss in the microwave frequency range of 8-12GHz. This synthesis

is needed for a radar wave absorber that can be absorbed in any frequency range, because we don't know what frequency is used by the opponent's radar.

CONCLUSION

The synthesis of $CoTiO_3$ material using the mechanical milling method showed that the phase in the XRD characterization test formed a single phase. SEM observation results show that the material is completely homogeneous. The results of the VSM data show that the material is formed superparamagnetic. The synthesis has succeeded in obtaining the value of reflection loss on the magnetic absorber material in the average X-Band frequency area.

Further research needed for calculating absorbed, reflected, and transmitted coefficient of electromagnetic wave by using analytical (use Maxwell equation and simulation) or experimental for data radar.

AUTHOR CONTRIBUTIONS

Each author of this article played an important role in the process of method conceptualization, and article writing.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

REFERENCES

Adi, W. A. (2010). Analisis Struktur dan Sifat Magnetik Paduan Sistem (BaSr)0.6(Fe(2(1-x)(MnTi)xO3 (x= 0; 0,25; 0,5), Tesis S2 Fakultas Matematika dan Ilmu Pengetahuan Alam, Universitas Indonesia.

Gaylor, K. (1989). Radar Absorbing Material-Mechanic and Materials. *DSTO Material Research Laboratory*, 1-23.

Jia, Z., Lan, D., Lin, K., Qin, M., Kou, K., Wu, G., & Wu, H. (2018). Progress in low-frequency microwave absorbing materials. *Journal of Materials Science in Electronic*, 29(2018), 17122-17136. https://doi.org/10.1007/s10854-018-9909-z

Jiang, J., Ai, L., Qin, D., Liu, H. & Li, L. (2008). Preparation and Characterization of Electromagnetic Functionalized Polyaniline/BaFe12O19 Composites. *Synthetic Metals*, 159(7-8), 695–699. https://doi.org/10.1016/j.synthmet.2008.12.021

Koutzarova, T.; Kolev, S.; Ghelev, C.; Grigorov, K.; & Nedkov, I. (2009). Structural and magnetic properties and preparation techniques of nanosized M-type hexaferrite powders. *Advances in Nanoscale Magnetism*, 122, 183–203. https://doi.org/10.1007/978-3-540-69882-1_10

Nasution, E.L, & Astuti. (2012). Sintesis Nanokomposit PAni/Fe3O4 sebagai Penyerap Magnetik pada Gelombang Mikro. *Jurnal Fisika Universitas Andalas*, 1(1), https://doi.org/10.25077/jfu.1.1.%25p.2012

Pullar, R.C. (2012). Hexagonal ferrites: A review of the synthesis, properties and applications of hexaferrite ceramics. *Progress in Materials Science.* p 57, 1191–1334. https://doi.org/10.1016/j.pmatsci.2012.04.001

Roya Bakhshkandi and Mahmood Ghoranneviss. (2019). Investigating the synthesis and growth of titanium dioxide nanoparticleson a cobalt catalyst. *Journal of Research In Science, Engineering And Technology*, 7(04), 1-3. https://doi.org/10.24200/jrset.vol7iss4pp1-3

Saptari, S. A. (2015). Sintesis dan karakterisasi sifat magnetik dan sifat listrik senyawa La0.67Ba0.33Mn1-xNix/2Tix/2O3 untuk aplikasi material penyerap gelombang mikro. Depok: Universitas Indonesia.

Weldon, D.G. (2009). Failure Analysus of Paints and Coatings. Wiley Online Library, USA. https://doi.org/10.1002/9780470744673