Microwave Absorber Sheet of the Composite Silicon Rubber – Iron Oxide

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Abstract. Microwave absorber sheet is a composite of silicon rubber – iron oxide. The composite was prepared by silicon rubber, toluene, and magnetic powder of iron oxide. The raw material was blended in the beaker glass for 60 min and then pressed at temperature of 70 °C for 15 min. The unsure analysis showed that the sample contained carbon, oxygen, sulfur, zinc, lanthanum, barium, manganese, titanium and iron. The refinement results of x-ray diffraction pattern show that the sample is semicrystalline with the crystallinity of 46%. The sample consist of the matrix is amorphous phase and the filler is crystalline phase. The filler consist of two phases, namely iron oxide and iron metal phases. The functional groups analysis show that the sulfur addition modified the polymer by forming crosslink (bridges) between individual polymer chains and bonding between magnetic filler and rubber matrix. We concluded that this study has been successfully made a composite of silicon rubber – iron oxide for microwave absorber sheet application.

1. Introduction
Information technology has become an important element in everyday life. The role of information technology in human activity at the present times is great and modern devices, such as mobile phones and computers are very complex and most of the electronic components are operating in the high frequencies range. However these electronic components are prone to leaks that resulted in the presence of interference frequency electromagnetic waves that can interfere with the performance of the equipment system [1]. Usually the electronic components mentioned above are mounted on a polymeric material that can absorb electromagnetic waves, also known as microwave absorber sheet. Microwave absorber sheet consists of a matrix made of polymer and filler material containing an electromagnetic wave absorbent material based upon magnetic materials. It means that the material is a composite that can absorb electromagnetic waves at a certain frequency range.

Fabricating absorbing materials provides an effective way to overcome these problems due to the dielectric loss or magnetic loss. Recently many authors have carried out studies on the absorbents and agents in order to improve absorbing properties of the composites [2–5]. It is demonstrated that a good absorbing material should satisfy two important conditions: 1) the intrinsic impedance of the absorbing materials is made to be equal to that of the free space; 2) the incident EM wave must enter and be rapidly attenuated through the material layer. Therefore, adjusting the complex permittivity and complex permeability of composites plays a key role in achieving excellent absorbing properties [6]. Presently, some research is concentrating on an attempt to modify certain magnetic materials such that the magnetic material can serve as an electromagnetic wave absorbent material [7–11]. One is the use...
of ferrite-based magnetic material, known as M-type hexagonal ferrite. This type of magnetic material is very promising because this material has some properties required as electromagnetic wave absorbent material. However, the synthesis of magnetic material of this kind is not easy to do and in order to meet the needs of mass production, there arise some constraints which result in a relatively high production cost. Beside that the development of electronic components is growing rapidly with a relatively cheaper price. Therefore, this study was conducted in order to find an alternative use of the product market of iron-based magnetic material and iron oxide. Iron particles (IPs) and iron oxide magnetic particle instant confirmation are used in absorbing sheet as an effective absorbent, they possess a good EM microwave absorption properties due to the large values of saturation magnetization and large Snokes limit in the gigahertz (GHz) frequencyrange.

The purpose of this work is to analyze the influence of silicone rubber composites filled with spherical iron/iron oxide composition on the microwave EM absorption properties. The main focus is on the effects ofweight content and shape of the iron and iron oxideunder silicone rubber on the reflection loss of the absorbing composites.

2. Materials and Methods

The absorber sheet in this study consists of silicon rubber, iron powder, and iron oxide. The matrix of absorber sheet were used was Methyl vinylsilicone rubber with toluene used as vulcanized assistants. The raw commercial of silicon rubber were supplied by best product, Germany, and raw commercial of iron oxide were supplied by pigment of local resources, Indonesia, which were prepared by a mechanical millingprocess using high energy millingmethod. The average particle diameter of the iron oxide was 100 – 300 nm and that of iron was 1 – 2 µm. The area of absorber sheet was about 25 mm2 and the thickness was about 1.5 mm. The 50% volume fraction of the iron and iron oxidewere added into the silicone rubber, andthen different amountof iron (the weight ratiowas 10wt%, 25wt% and 50 wt% relative to the iron oxide weight) was mixed in each composite. The three synthesized samples, each with a with different composition are shown in Table 1.

| No. | Sample | Silicon rubber | Pigment (iron oxide) | Iron |
|-----|--------|----------------|---------------------|------|
| 1.  | Sheet-1| 50             | 40                  | 5    |
| 2.  | Sheet-2| 50             | 37.5                | 12.5 |
| 3.  | Sheet-3| 50             | 25                  | 25   |

The silicone rubber and absorbents were mixed according to two procedures. Firstly the iron oxidewas added to the silicone rubber to guarantee a better dispersion. Secondly the iron was added to the previous compounds. The preparation result of absorber sheet is shown in figure 1.

![Figure 1. Sample of absorber sheet](image)

The surface morphology- and elemental composition examination of the sample was carried out by using the JEOL scanning electron microscope (SEM) and energy dispersive spectroscopy (EDS) method, respectively. The phase qualitative and quantitative analysis were carried out using the Pan
Analytical diffractometer equipped (XRD) with a tube provided with a copper anode. The wavelength of X-ray radiation (CuKα) was 1.5406 Å. The phase analysis was performed by using GSAS program (Rietveld method) (Toby, 2001). The analysis of functional groups was carried out employing the Fourier transformation infra red (FTIR) method. The magnetic properties were evaluated using an Oxford instrument vibrating sample magnetometer (VSM) apparatus. Finally, the reflection loss of microwave was carried out using the vector network analyzer (VNA) with a frequency range of 300 kHz - 20 GHz.

3. Results and Discussion
Figure 2 shows the SEM image of absorber sheet which prepared by the mixture of silicon rubber elastomer thermoplastics, pigment (iron oxide), and iron powder.

In figure 2 it appears that the distribution of the filler in the form of iron oxide pigments in the silicone rubber matrix looks very homogeneous, in the form of polymer sheets, which is excellent. However, after being added to the filler metal, the composite looks like scattered granules, scattered over the surface of the sheet. The more filler added more iron granules are scattered over the surface of the sheet while the homogenization level decreases. This is presumably due to the higher density level composite, which makes the homogenization of the filler into the polymer matrix more difficult.
Figure 3 is showed the results of the elementary analysis by using energy dispersive spectroscopy on the filler and matrix of the composite material.

![Figure 3](image1.png)

(a) Filler

![Figure 3](image2.png)

(b) Matrix

**Figure 3.** The elementary analysis of composite material by using energy dispersive spectroscopy

The energy dispersive spectrum as in figure1 shows that the dominant elements of the filler are carbon (C), and iron (Fe) while the dominant elements of the matrix are carbon (C), oxygen (O), silicon (Si), iron (Fe), and copper (Cu). The detailed content of the sample are showed in Table 2.

| No. | Unsure          | Normalize of Content (wt%) | Filler | Matrix   |
|-----|----------------|----------------------------|--------|----------|
| 1.  | Copper (Cu)    | -                          | 1.32 ± 0.94 |          |
| 2.  | Iron (Fe)      | 79.68 ± 1.60               | 22.36 ± 1.72 |        |
| 3.  | Silicon (Si)   | -                          | 23.85 ± 1.59 |        |
| 4.  | Oxygen (O)     | -                          | 7.49 ± 2.83 |        |
| 5.  | Carbon (C)     | 20.32 ± 0.65               | 44.98 ± 1.44 |        |

**Table 2.** The result of elementer analysis of the composite

Microstructure examination of the sample shows that the filler of magnetic particles are embedded in the silicon rubber matrix and are distributed on the sample’s surface. Based on the results of the elemental analysis it was found that iron powder as filler contained in the composite have materialized. So that, it is needed a further confirmation about phases formed in the sample by using XRD as shown in figure 4.
The refinement result of XRD pattern on the samples has produced very good quality of fitting with R factor being very small. And goodness of fit value $\chi^2$ (chi-squared) is found to be in agreement with the Rietveld standard [14]. The sample is semicrystalline with the crystallinity of 50%. And the sample consist of the matrix was amorphose phase and the filler was crystalline phase. The filler consist of four phases, namely CuFeS$_2$ (chalcopyrite), FeS$_2$ (pyrite), FeO$_2$ (goethite) and Fe(iron) phases. The chalcopyrite phase had tetragonal structure, space group of $I-42d$ (COD-1010940), pyrite phase had cubic structure, space group of $Pa-3$ (COD-9006170), goethite phase had orthorombic structure, space group of $Pbnm$ (COD-9016406), and iron phase had cubic structure, space group of $Im-3m$ (COD-9013415). This result is evidenced by the calculation results of the mass fraction as shown in Table 3.

Table 3. Mass fraction of the samples

| Sample   | Phase    | Mass fraction (%) | Rwp | $\chi^2$ (chi-squared) |
|----------|----------|-------------------|-----|------------------------|
| Sheet-1  | CuFeS$_2$| 5.89(5)           |     |                        |
|          | FeS$_2$  | 2.16(1)           |     |                        |
|          | FeO$_2$  | 72.69(1)          |     |                        |
|          | Fe       | 19.24(2)          | 2.77| 1.135                  |
| Sheet-2  | CuFeS$_2$| 3.97(3)           |     |                        |
|          | FeS$_2$  | 1.08(8)           |     |                        |
|          | FeO$_2$  | 65.86(1)          |     |                        |
|          | Fe       | 32.09(9)          | 2.38| 1.160                  |
| Sheet-3  | CuFeS$_2$| 1.98(4)           |     |                        |
|          | FeS$_2$  | 1.16(9)           |     |                        |
|          | FeO$_2$  | 43.32(1)          | 2.70| 1.182                  |
|          | Fe       | 53.53(1)          |     |                        |
However, the XRD result could not explain the bonding mechanism including the formation of crosslink (bridges) between individual polymer chains in the composite, so that additional confirmation is needed by using FTIR equipment as shown in figure 5.

![FTIR spectrum](image)

**Figure 5.** The FTIR transmittance spectrum of the sample.

The vibrational spectrum of a molecule is considered to be a unique physical property of the molecule and is characteristic of a molecule. A quantitative analysis of structure and confirmation of polymer chains requires the assignment of absorption bands and qualitative measurement of their intensities. The assignments of the fundamental frequencies have been made on the basis of magnitude and relative intensities of the observed bands. The vibrational band assignments of the rubber materials have been made in correlation with molecular composition and with their indices of stereo regularity. Figure 5 represented the FTIR spectrum between 4000 to 600 cm$^{-1}$ of the composite sample. Transmittance peaks of the FTIR spectra on the composite sample indicate the presence of vibration of O–H, C–H, Si–C, Si–O, and Fe–O bond [12]. Natural rubber consists of suitable polymers of the organic compound isoprene with minor impurities of other organic compounds and water. According to the functional groups analysis by using FTIR showed that transmittance peaks of the silicon rubber appeared at wave numbers around 3000 cm$^{-1}$, 1250 cm$^{-1}$, and 1050 – 750 cm$^{-1}$ that indicated the presence of the C–H, Si–C, and Si–O bond vibrations, respectively. However the transmittance peaks of the iron oxide also observed at wave numbers around 3700 cm$^{-1}$ and 600 cm$^{-1}$ that indicate the presence of the H–O and Fe–O bond vibrations, respectively. It is suspected that there was bonding between iron oxide as filler and silicon rubber as matrix.

Characterization of the magnetic properties of the absorber sheet was performed using a vibrating sample magnetometer (VSM) that produced hysteresis curves of magnetic particles.

![Hysteresis curves](image)

**Figure 6.** The hysteresis curves of absorber sheet
The hysteresis curves of synthesized absorber sheet are shown in figure 6 in which respective profiles for sheet-1, sheet-2, and sheet-3 are also compared. The hysteresis loop as shown in figure 6 is characterized with intrinsic saturation $M_s$, remanence field $M_r$ and coercivity $H_c$. The intrinsic saturation, $M_s$ is the state when the material is not able to absorb more magnetic field, so increasing magnetization force will not change magnetic flux density significantly. Meanwhile, the remanence field, $M_r$ is the residual magnetization in a medium once the external magnetic field is removed and coercivity (also called coercive force) is a force required to demagnetize the material so the residual induction becomes zero after magnetizing up to saturation.

![Figure 6. Hysteresis curves of synthesized absorber sheet](image)

**Figure 7.** The reflection loss (RL) curve of absorber sheet

The microwave absorbing properties of synthesized absorber sheet is shown in figure 7 in which respective profiles for sheet-1, sheet-2, and sheet-3 are also compared. Figure 4 shows the relation between the reflection loss (RL) of absorber sheet and the microwave frequency X-band in the range of 10-15 GHz when the thickness of the sample is 1.5 mm. There are at least a absorption peaks observed which have a high RL within the frequency range for all samples. Base on the phase composition show that the absorber sheet has a low RL which then increases. This case shows that however the best absorber sheet is occurred at nearly the same thickness and at practically identical frequencies, there are differences. This can be explained by the effect of electromagnetic properties on the attenuation characteristic in each sample. This indicates that absorber sheet has certain microwave absorption properties in the frequency range of 10 – 15 GHz, absorption peak values of -15 dB at 12 GHz.

### 4. Conclusion

The synthesis and characterization of absorber sheet have been carried out using chemical process. Microstructure of the sample showed that the filler of magnetic particles embedded in the silicon rubber matrix and distributed homogeneously on the sample surface. The unsure analysis showed that the sample contained carbon (C), oxygen (O), silicon (Si), iron (Fe), and copper (Cu). The refinement results of x-ray diffraction pattern showed that the sample is semicrystalline with the crystallinity of 50%. And the sample consist of the matrix was amorphose phase and the filler was crystalline phase. The filler consist of two phases, namely CuFe$_2$S$_2$ (chalcopyrite), FeS$_2$ (pyrite), FeO$_2$ (goethite) and Fe(iron) phases. The functional groups analysis showed that the sulfur addition modified the polymer by forming crosslink (bridges) between individual polymer chains and bonding between magnetic filler and rubber matrix. This study has been successfully synthesized silicon rubber – iron oxide and has been understood changes in the microstructure, crystal structure and bonding mechanism of the composite.
Acknowledgements
This work is supported by the program for research and development of smart magnetic material (DIPA 2015), Center for Science and Technology of Advanced Materials, National Nuclear Energy Agency. The authors are thankful to Drs. Gunawan, M.Sc. and Edy Giri R.P., Ph.D., for their permission to publish this paper. Many thank to Dra. Mujamilah, M.Sc. for their kind help to characterize using VSM.

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